

MANAGEMENT & OPERATIONS

Evaluating GAC for Trihalomethane Control

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The primary objectives of this study were to optimize granular activated carbon (GAC) treatment to meet more stringent trihalomethane (THM) standards, to estimate the costs of optimized GAC treatment, to identify by-products of the final disinfection of GAC-treated water, and to assess the ability of the rapid small-scale column test (RSSCT) to simulate total organic carbon (TOC) breakthrough from pilot-scale carbon contactors. The lowest THM concentration achievable by means of GAC treatment was found to be 10 $\mu\text{g/L}$. It was concluded that this type of treatment is expensive for the control of THMs, the predominant by-products of postdisinfection of GAC effluents with chlorine or chloramines. The RSSCT was shown to be an economical means of estimating pilot-scale TOC breakthrough, which can be used in making preliminary estimates of carbon utilization rates and cost estimates.

The 1986 amendments to the Safe Drinking Water Act identify granular activated carbon (GAC) as a feasible treatment technique for synthetic organic chemicals.¹ Trihalomethanes (THMs) may be included among the compounds for which GAC could be considered a feasible treatment technology. The current drinking water

standard for total THMs is 0.10 mg/L (100 $\mu\text{g/L}$). Several recent actions by the US Environmental Protection Agency (USEPA) suggest that this maximum contaminant level (MCL) for THMs will be lower in the future.

On Jan. 22, 1988, the USEPA issued a final priority list of disinfection by-products (DBPs), including THMs.² Proposed

MCL goals and actual MCLs are expected to be issued in the early 1990s. At the present time, it is not known what these MCLs will be. The USEPA may choose to regulate THMs on an individual basis rather than combined as a total of the four compounds. The MCL could be as low as 5 $\mu\text{g/L}$ and probably not higher than 50 $\mu\text{g/L}$ (for a total of the four THMs). The THM treatment objectives assessed as part of this study were 5, 10, 20, and 50 $\mu\text{g/L}$; these goals were selected because they span the range of possible new THM regulations.

The purpose of this article is to present the significant findings of a study entitled "Optimization and Economic Evaluation of Granular Activated Carbon for Organic Removal."³ The full report of this study and other related publications are cited throughout this article and should be consulted for important methodological details.

Objectives

The primary objectives of this study, which were directed toward the needs of the Metropolitan Water District of Southern California (MWD) and the water industry as a whole, were

- to optimize GAC treatment for meeting lower THM standards,
- to estimate the costs associated with optimized GAC treatment for MWD and for six other water utilities across the United States,
- to identify DBPs resulting from the final disinfection of GAC-treated water with chlorine or chloramines,
- to assess the ability of a mathematical model or bench-scale column test such as the rapid small-scale column test (RSSCT) to simulate total organic carbon (TOC) breakthrough curves from pilot-scale carbon contactors, and
- to develop helpful information and evaluation techniques that can be used

A full report of this project (catalog no. 90550) is available from the AWWA Research Foundation, 6666 W. Quincy Ave., Denver, CO 80235.

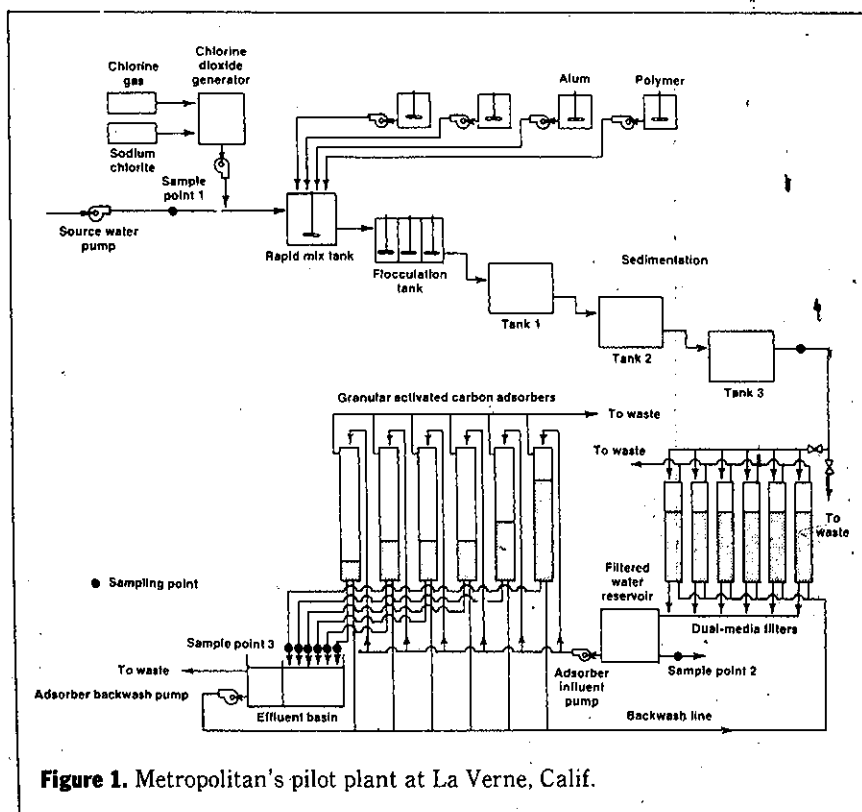


Figure 1. Metropolitan's pilot plant at La Verne, Calif.

by other utilities in evaluating GAC treatment for their facilities.

Scope of work

The scope of work for this study involved

- operating six postfiltration GAC contactors, with different empty bed contact times (EBCTs) and GAC mesh sizes, to develop breakthrough profiles for the following: TOC, THMs, total organic halogen (TOX) compounds, other DBPs (including trichloroacetic acid [TCAA], dichloroacetic acid [DCAA], and dihaloacetonitriles [DHANs]), and simulated distribution system (SDS) THMs, TOC compounds, TCAA, DCAA, and DHANs associated with postchlorination and postchloramination;

- evaluating the impact of GAC treatment on the microbial quality of the treated water;

- developing cost estimates associated with GAC treatment for SDSTHM treatment goals of 5, 10, 20, and 50 µg/L;

- using mathematical models to simulate the pilot-column TOC breakthrough curves to predict the optimum EBCT;

- using an RSSCT to simulate TOC breakthrough profiles from pilot-scale carbon contactors; and

- performing an RSSCT for six other US water utilities, measuring both the TOC and SDSTHM breakthrough profiles, so that cost estimates associated with GAC treatment could be prepared for these utilities.

Background

Metropolitan's treatment facilities. Metropolitan's treatment system, which includes five treatment facilities that will be expanded to a capacity of almost 3 bgd by the year 2000, is one of the largest in the world and supplies most of the treated water used in Southern California. The source waters for this system are Colorado River water (CRW) and state project water (SPW). The Robert A. Skinner, Robert B. Diemer, and F. E. Weymouth filtration plants can use both sources. The Henry J. Mills Filtration Plant uses only SPW from the East Branch of the California Aqueduct, and the Joseph Jensen Filtration Plant uses only SPW from the West Branch. The process train employed at most of the plants includes predisinfection with chlorine, coagulation, flocculation, sedimentation, filtration, pH adjustment with sodium hydroxide, and final disinfection with chloramines. All of these plants are to be expanded within the next seven to ten years. Typical water quality characteristics of CRW and SPW are summarized in Table 1.

Pilot treatment facility. The pilot treatment facility (Figure 1) is located at MWD's Weymouth plant in La Verne, Calif. It is a 6-gpm facility, constructed entirely of glass, stainless steel, and

TABLE 1
Typical water quality of CRW and SPW during MWD's GAC pilot studies

Source Water	Water Quality Parameter	Concentrations Measured During Study		
		Mean	Maximum	Minimum
CRW	Temperature—°C	12.5	15.0	11.5
	Turbidity—ntu	1.6		
	Conductivity—µmho/cm	805	854	400
	Bromide—mg/L	0.06	0.12	0.05
	Total alkalinity—mg as CaCO ₃ /L	129		
	TOC—mg/L	2.64	2.97	2.31
	TOX—µg/L	25	40	12
SPW	Temperature—°C	24.0	25.0	22.5
	Turbidity—ntu	2.3		
	Conductivity—µmho/cm	474	520	452
	Bromide—mg/L	0.17	0.19	0.14
	Total alkalinity—mg as CaCO ₃ /L	76		
	TOC—mg/L	2.52	2.82	2.27
	TOX—µg/L	19	29	12

TABLE 2
Analytical methods

Parameter	Method	Reference
THMs	LLE-GC-ECD; USEPA method 501.2	3
DCAA-TCAA	LLE(derivatization)-GC-ECD	3
DHANs	LLE(salt)-GC-ECD	3
SDS	1.0 mg Cl ₂ /L, 1.5 mg chloramines/L, pH = 8.2, five days, 25°C	3
TOX	SW 846 method 9020	3.5
Free-combined chlorine	DPD method, DR1/A colorimeter	3.6
Chlorine dioxide, chlorite, chlorate	Amperometric titration	3.7
TOC	Persulfate-ultraviolet oxidation	3.6
Bromide	Ion chromatography	3
UV absorbance	Measured at 250 nm, Lambda 5, Perkin Elmer UV spectrophotometer	3
pH, turbidity, specific conductance	Standard Methods	3.6

TFE-FEP, and it is plumbed to receive 100 percent CRW, 100 percent SPW, or any blend of these two waters. The treatment train includes predisinfection (with chlorine dioxide), coagulation-flocculation (using alum and a cationic polymer), sedimentation, dual-media filtration, six GAC columns (operated in parallel), and laboratory-simulated post-chlorination or postchloramination. The GAC column EBCTs tested during this study were 7.5 min, 15 min, 30 min, and 60 min. A commercial (12×40 mesh) size GAC* was used for the pilot-scale studies reported here.

The breakthrough profile data obtained from each of the four carbon EBCTs are primarily representative of CRW. However, after approximately three months of pilot-plant operation, the source water was switched to a blend of CRW and SPW as a result of operational changes at the Weymouth plant. Limited data are presented on TOC and SDSTHM breakthrough for column 4, which only received SPW late in the study.

Methods

RSSCT. The rapid small-scale column is a small-scale replica of a pilot- or full-

scale fixed-bed carbon adsorber and is capable of producing breakthrough profiles similar to those from a larger-scale contactor. It uses a much smaller carbon particle size and carbon bed size and requires much less water and time to perform than its larger-scale counterpart. As part of this study, RSSCTs were performed to simulate the TOC breakthrough profiles from the 15-, 30-, and 60-min-EBCT pilot-scale carbon contactors.

The experimental setup and column packing for the RSSCT technique are shown in Figure 2. The 12×40 mesh carbon was crushed and sieved to the desired particle size. A previous article by Crittenden⁴ describes the RSSCT method in some detail. Experience gained in this study suggests that the cost to complete one RSSCT is about one tenth to one hundredth of the cost of a pilot-scale test, depending on the complexity of the pilot study.

Analyses. Table 2 summarizes the analytical methods used.^{3,5-7} Where appropriate, outside references are listed. However, many of the methods were developed or modified specifically for

*Filtrisorb 400, Calgon Carbon, Pittsburgh, Pa.

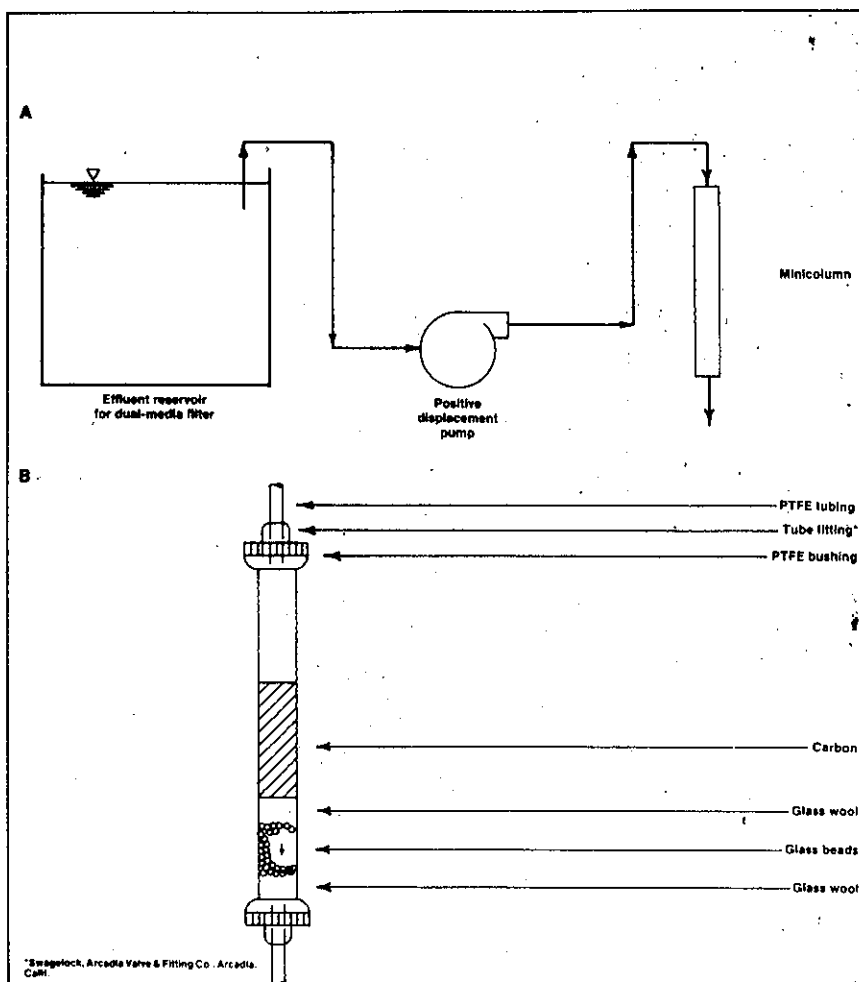


Figure 2. Setup for RSSCT (A) and minicolumn packing (B)

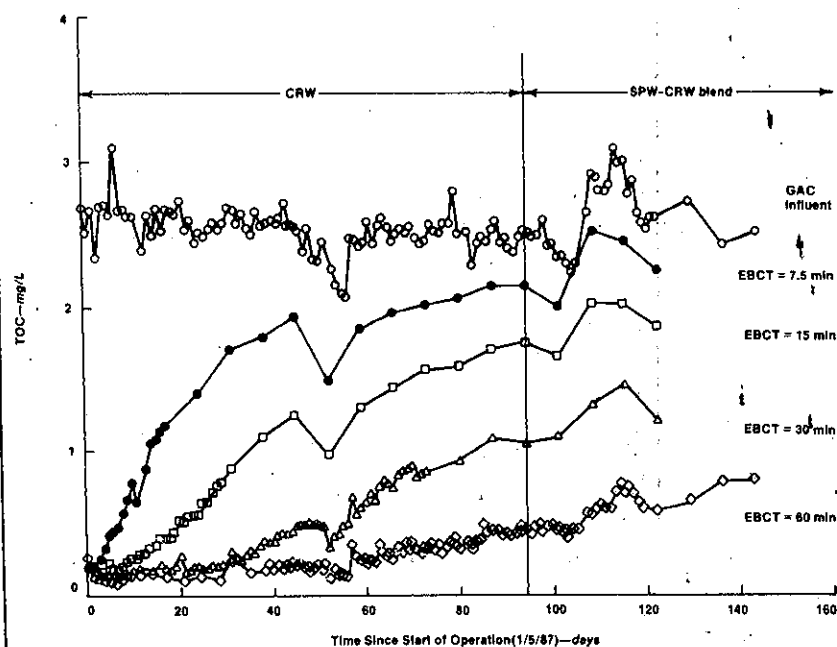


Figure 3. Pilot GAC column influent TOC concentrations and TOC breakthrough profiles for MWD

this project, and these are available in Appendix A of the full report.³

The SDS technique was used during this study to simulate, in the laboratory, DBP concentrations that would be expected to occur in MWD's distribution system as a result of postdisinfection. The samples were spiked with either 1.0 mg free chlorine/L or 1.5 mg chloramines/L and incubated at 25°C for five days. The incubation temperature, 25°C, was selected because it is representative of the relatively high temperatures in MWD's distribution system. The incubation time, five days, was selected because it approximates the maximum detention time through MWD's distribution system. The disinfectant dosages and the incubation temperature and time used for this SDS technique are specific to MWD's system and may not be applicable to other utilities' distribution systems. The subscripts "Cl" and "NH" are used to distinguish between samples spiked with chlorine and chloramines, respectively.

Results from MWD's pilot plant

Breakthrough profiles. The TOC and SDSTH_{Cl} breakthrough profiles for



An engineer at the Metropolitan Water District of Southern California loads the rotating tumbler for agitating the RSSCT samples.

the different EBCTs tested are shown in Figures 3 and 4, respectively. Although breakthrough profiles were developed for other DBPs, those for TOC and $\text{SDSTHM}_{\text{Cl}}$ are included in this article because they were the ones that were used in preparing the cost estimates for GAC treatment. Breakthrough profiles for TOX are discussed later in this section. THMs were observed to be the predominant DBPs formed during post-disinfection. Because THMs predominated, using GAC to control THM concentrations should, at the same time, control the formation of other DBPs. Using the breakthrough profiles presented in Figures 3 and 4 (which represent essentially 100 percent CRW), the regeneration frequencies and effluent TOC concentrations corresponding to the different $\text{SDSTHM}_{\text{Cl}}$ treatment goals were determined (Table 3).

Figure 5 shows, in more detail, the breakthrough of the $5\text{-}\mu\text{g/L}$ nonadsorbable $\text{SDSTHM}_{\text{Cl}}$ fraction during the first

few days of operation for all four EBCTs. Although this phenomenon has been noted by other researchers, its importance in meeting very low THM standards should be emphasized. If this phenomenon is confirmed in other GAC studies of precursor removal, it will limit the lower level of a future THM standard to about $10\text{ }\mu\text{g/L}$.

The GAC in column 4 (EBCT = 15 min) was replaced with virgin carbon four times over the course of the study. As a result, separate breakthrough profiles were obtained from column 4 for 100 percent CRW, 100 percent SPW, and a blend of the two waters. By comparing

these curves, shown in Figures 6 and 7, the following observations can be made:

- little difference was seen between the TOC breakthrough curves for 100 percent CRW and the CRW-SPW blend,
- the TOC breakthrough for 100 percent SPW occurred at a slightly slower rate than did TOC breakthrough for 100 percent CRW, and
- the THM precursor breakthrough was more rapid when treating 100 percent SPW than when treating 100 percent CRW. That is, at essentially the same TOC concentration, SPW exhibited a higher potential for THM formation than did CRW. This is presumably the result

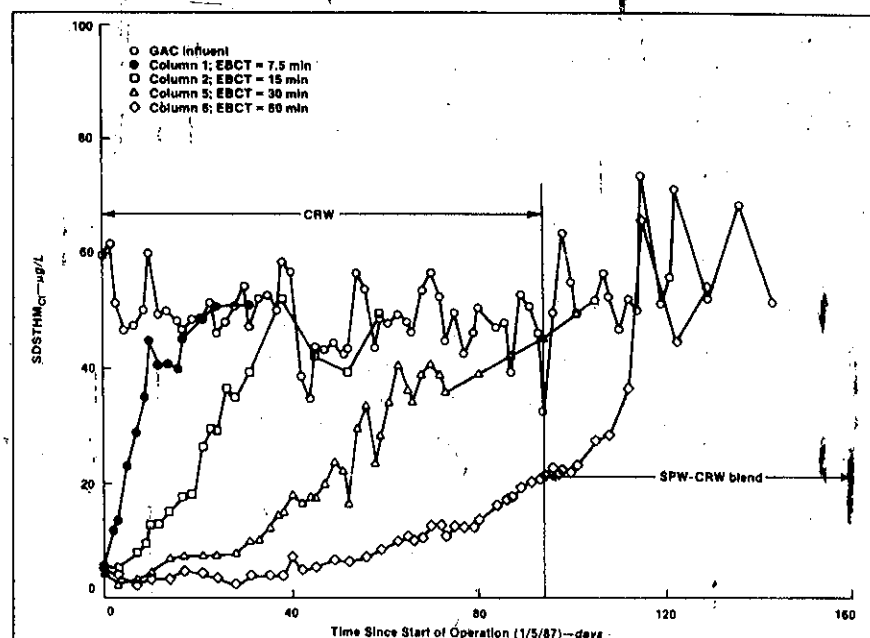


Figure 4. Pilot GAC column influent $\text{SDSTHM}_{\text{Cl}}$ concentrations and $\text{SDSTHM}_{\text{Cl}}$ breakthrough profiles for MWD

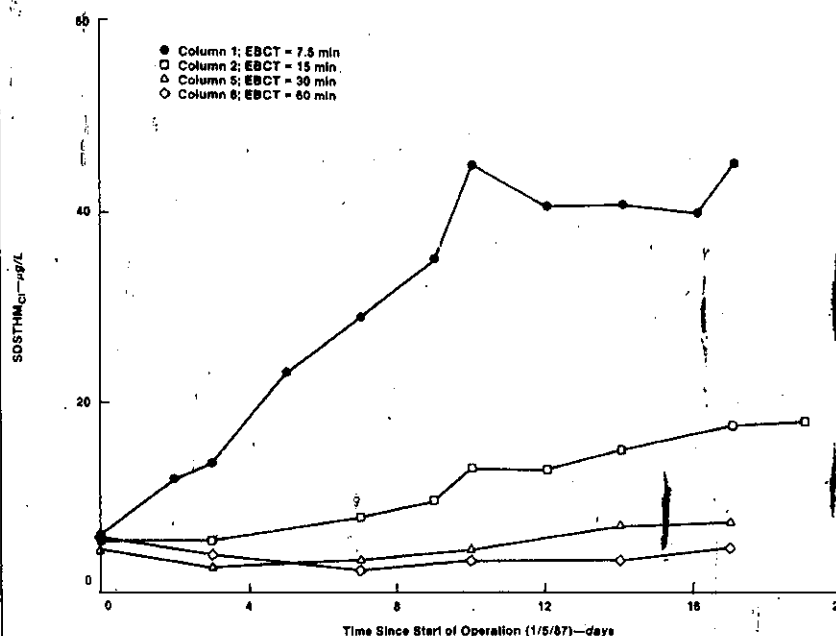


Figure 5. Pilot GAC column $\text{SDSTHM}_{\text{Cl}}$ breakthrough profiles for first 20 days of column operation for MWD

of SPW's higher bromide concentration and the presence of a different type of precursor than that found in CRW.

The GAC regeneration frequencies for the treatment of 100 percent SPW are reported in Table 4.

THM formation. Postdisinfection of GAC-treated water with chlorine was shown to produce substantially higher SDSTHM concentrations than postdisinfection with chloramines. Comparing the SDSTHM_{Cl} breakthrough curves in Figure 4 and the SDSTHM_{NH} breakthrough curves shown in Figure 8 illustrates this point. Very small concentrations—<4 µg THMs/L—broke through, even during the shortest EBCT. However, because of the microbiological concerns about GAC column effluents, it is unlikely that the use of chloramines as a primary disinfectant would be prudent downstream from GAC filters. However, a combination of a short contact time with free chlorine followed by conversion to chloramines could be a workable scenario.

TOC as a surrogate. Other researchers^{8,9} have evaluated the use of surrogate parameters as indicators of other water quality parameters that are more difficult to measure, and they have shown that TOC can be used successfully as an indicator of THM formation potential. The TOC serves as a good indicator of SDSTHM_{Cl} because a significant portion of the organic compounds that constitute TOC are potential precursors of THM compounds. The SDSTHM_{Cl} analysis is much more difficult and requires substantially more time to perform than the TOC analysis. In a full-scale application, it would be impractical and extremely difficult to routinely monitor SDSTHM_{Cl} in order to determine when the appropriate time for carbon replacement or regeneration would be. The results of this study also demonstrate that ultraviolet (UV) absorbance is a suitable surrogate for TOC and, therefore, for SDSTHM_{Cl} (Figure 9). Because UV absorbance can be continuously monitored, it could be used, in a full-scale application, as an operational control parameter for SDSTHM_{Cl} breakthrough.

The results in Figure 10 show that TOC will serve as an excellent indicator of SDSTHM_{Cl} for CRW (provided that there is a free chlorine residual remaining in the SDS sample at the end of the incubation period). For both waters, the relationship between the two parameters is linear and can be described by the following equations (for this study only):

$$\begin{aligned} \text{CRW: } \text{SDSTHM}_{\text{Cl}} &= 58.89(\text{TOC}) - 2.30 \\ &\text{(valid for } 0 \leq \text{TOC} \leq 0.75) \\ r^2 &= 0.90 \end{aligned}$$

$$\begin{aligned} \text{SPW: } \text{SDSTHM}_{\text{Cl}} &= 97.41(\text{TOC}) - 6.36 \\ &\text{(valid for } 0 \leq \text{TOC} \leq 0.70) \\ r^2 &= 0.96 \end{aligned}$$

TABLE 3
TOC concentrations and GAC regeneration frequencies corresponding to different SDSTHM goals (Colorado River water)

SDSTHM _{Cl} Goal µg/L	TOC mg/L	GAC Regeneration Frequency—days			
		7.5-min EBCT	15-min EBCT	30-min EBCT	60-min EBCT
5	0.16	0	3.1	10	32
10	0.25	2.8	9	30	56
20	0.44	5.5	19	42	90
50	1.00	13.8	35	83	

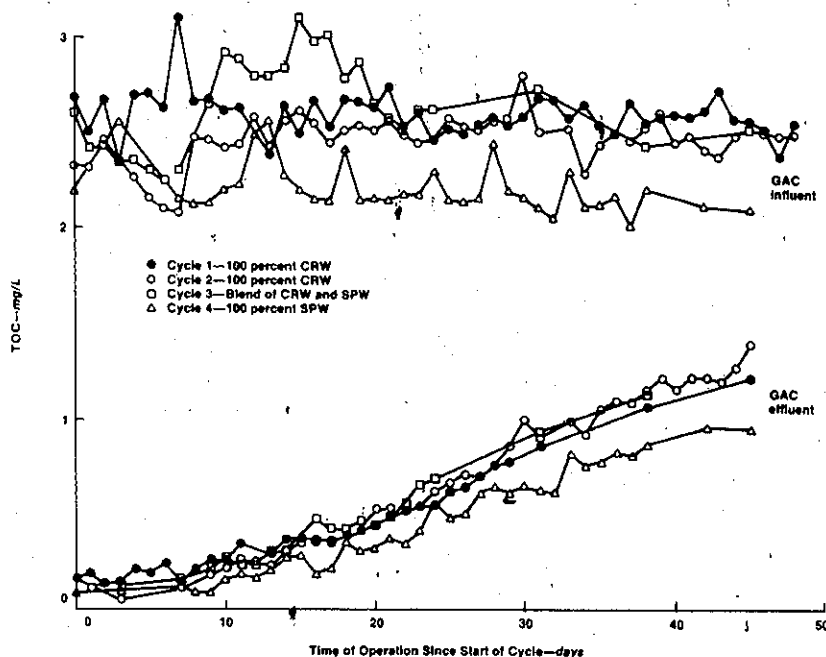


Figure 6. Pilot GAC column TOC breakthrough profiles for MWD (column 4; EBCT=15 min)

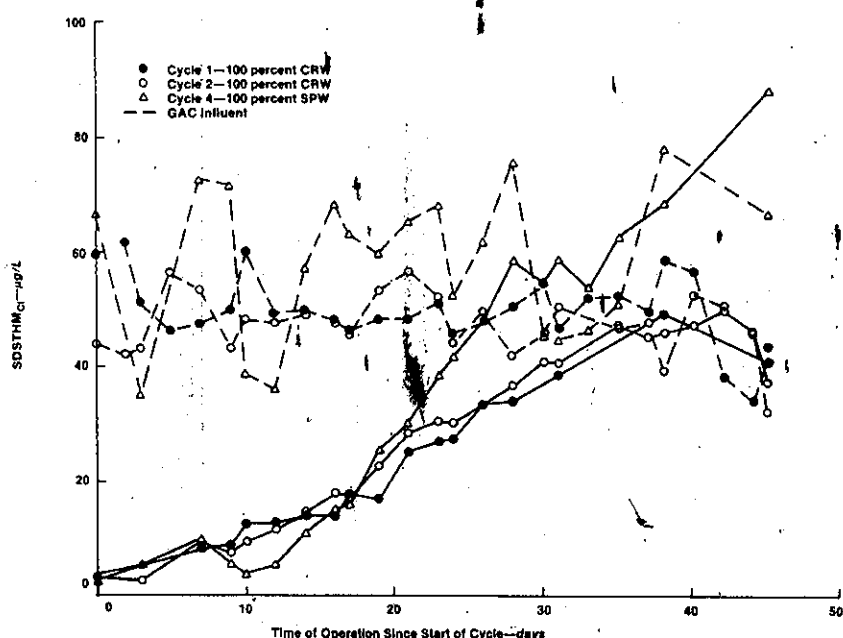


Figure 7. Pilot GAC column SDSTHM_{Cl} breakthrough profiles for MWD (column 4; EBCT=15 min)

TABLE 4
TOC concentrations and GAC regeneration frequencies corresponding to different SDSTHM goals (state project water)

SDSTHM _{Cl} Goal µg/L	TOC mg/L	GAC Regeneration Frequency (EBCT = 15 min) days
5	0.12	2
10	0.17	10
20	0.27	18
50	0.58	26

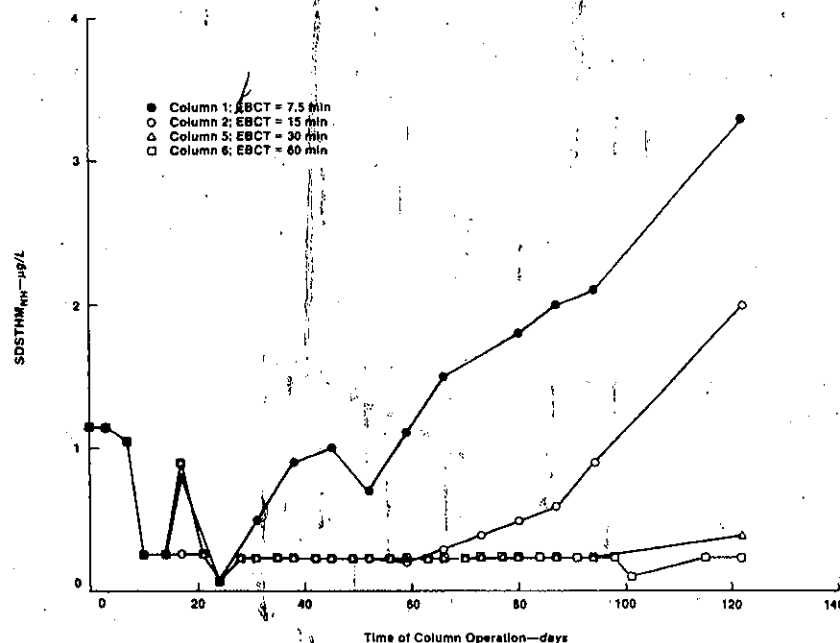


Figure 8. Pilot GAC column SDSTHM_{NH} breakthrough profiles for MWD

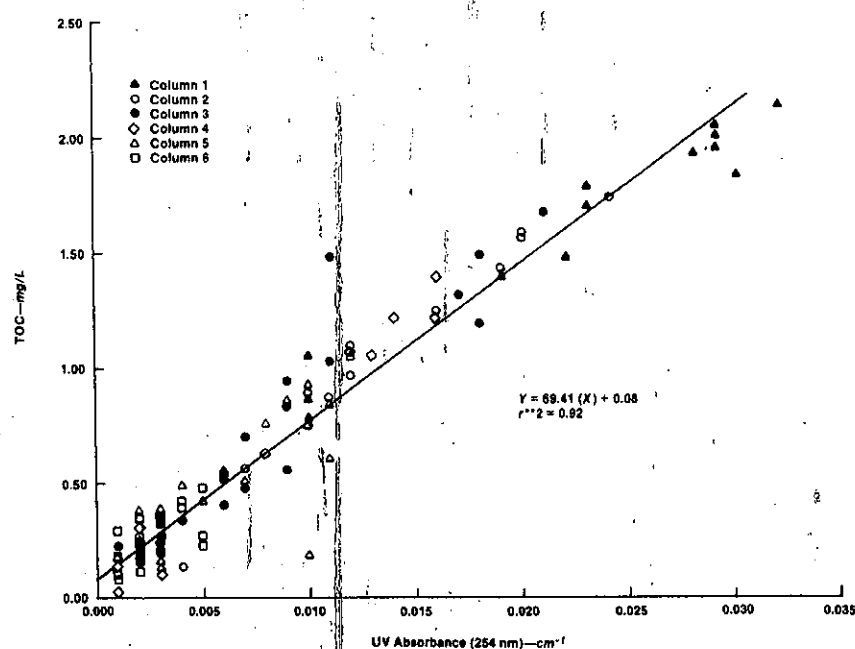


Figure 9. Linear relationship between TOC and UV absorbance (100 percent CRW)

in which the concentrations for SDSTHM_{Cl} and TOC are expressed in micrograms and milligrams per litre, respectively.

It is important to note that this relationship will be different for different waters and must be established for the specific water of interest. Depending on the organic matrix of the water, this relationship may vary with the season, time, or both.

TOX. The SDSTOX_{Cl} breakthrough profiles for the four different EBCTs examined in this study are presented in Figure 11. It is clear that these breakthrough curves are not nearly as smooth as those for TOC and SDSTHM_{Cl}. It is presumed that this is because of the inherent variability associated with the TOX analysis. With regard to EBCT, the SDSTOX_{Cl} breakthrough profiles follow the expected trend of increasing time to breakthrough with increasing contact time.

Comparison of RSSCT and pilot results

The scaling equations used to set up the RSSCT are based on a dimensional analysis of the equations included in the dispersed-flow, pore-surface diffusion model,¹⁰ which is used to simulate adsorbate breakthrough from a full-scale contactor. This model allows for similarity between the small- and large-scale breakthrough profiles because it addresses the fundamental mechanisms driving the adsorption process in a fixed-bed system: advective flow, axial dispersion and diffusion, liquid-phase mass transfer resistance, surface diffusion, pore diffusion, adsorption equilibrium, and competitive equilibrium of solutes on the carbon surface. Dimensional analysis of the equations in the computer model results in six independent, dimensionless groups. To achieve similarity between the RSSCT and any full-scale adsorber, each of the six dimensionless groups must remain constant as the full-scale adsorber is scaled down. The scaling equations for sizing the RSSCT were obtained by setting the dimensionless groups for the large-scale adsorber equal to those for the small-scale contactor. As has been discussed by Crittenden et al.,⁴ equating these dimensionless groups results in three sets of scaling equations, which depend on the functional relationship between the solute (in this case TOC) intraparticle diffusivity (surface and pore diffusivity) and the GAC particle size:

- set 1—intraparticle diffusivity is independent of particle size;
- set 2—intraparticle diffusivity is a linear function of particle size; and
- set 3—intraparticle diffusivity is related to particle size, but by some other functionality.

The first step in performing the RSSCTs for this study was to select the

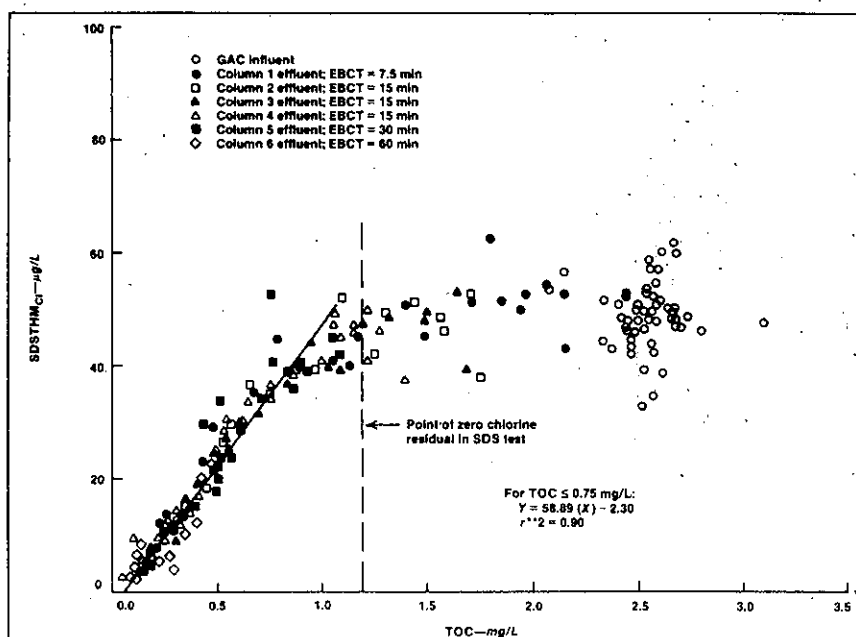


Figure 10. Linear relationship between $SDSTHM_{C1}$ and TOC (100 percent CRW)

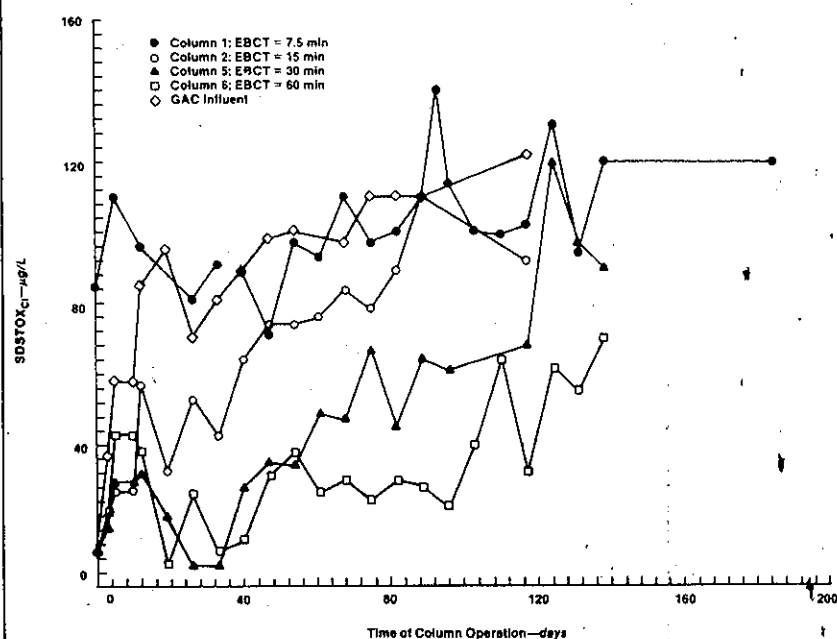


Figure 11. Pilot GAC column $SDSTOX_{C1}$ breakthrough profiles for MWD

appropriate scaling equations to be used in determining the correct mass of GAC and the surface loading rate for the RSSCT. Prior to the RSSCTs, a series of batch rate tests was conducted with CRW and SPW. These indicated that, for both waters, surface diffusivity was a linear function of particle size.

A comparison between the RSSCT-simulated TOC breakthrough profile and the pilot-scale breakthrough profile (100 percent CRW) for the 15-min-EBCT contactor is shown in Figure 12. The breakthrough curves are presented on the basis of "equivalent days of pilot column operation," which takes into account the scaling relationships used in

sizing the RSSCT. The fit between the two curves (in qualitative terms) is excellent. For the 30- and 60-min-EBCT columns (results not shown), the RSSCT and pilot breakthrough curves track one another at the lower effluent TOC concentrations, but the curves begin to deviate from one another at higher TOC concentrations. The fit between the RSSCT and pilot-scale curves (EBCT = 15 min) for 100 percent SPW was also good, but not as good as that for 100 percent CRW.

To provide a national perspective to the study, six utilities from across the United States were selected to participate in this project. RSSCTs were performed

for these six water utilities in order to estimate the costs associated with using carbon adsorption as a means of THM precursor control for waters of different qualities. The following utilities agreed to participate in this study and supplied MWD with 60 gal of untreated water for the test: Atlanta (Ga.) Water Bureau, Cincinnati (Ohio) Water Works, East Bay Municipal Utility District (Oakland, Calif.), Jefferson Parish (La.) Water District, Palm Beach County (Fla.) Water Utilities, and Philadelphia (Pa.) Water Department. Three of these six utilities—Cincinnati, Jefferson Parish, and Philadelphia—had previously performed pilot-scale GAC adsorption studies. As a means of validating the accuracy of the RSSCTs, TOC breakthrough profiles obtained from those earlier pilot studies were compared with the RSSCT-simulated breakthrough curves.

The RSSCT curves are compared with the pilot-scale curves for Cincinnati, Jefferson Parish, and Philadelphia in Figures 13, 14, and 15, respectively. As shown, good to excellent fits were attained for Cincinnati and Jefferson Parish. The Cincinnati pilot-plant TOC data, shown in Figure 13, were taken from a pilot study performed in the same season as that in which the RSSCT was performed. These pilot data, however, were collected in 1980, whereas the RSSCT data were collected in 1987. The two curves for Philadelphia do not compare well. A satisfactory RSSCT breakthrough curve was developed for Palm Beach, but no pilot data were available for comparison. As a result of the low concentrations of TOC in Atlanta's and East Bay's waters, TOC breakthrough profiles could not be developed with the available 60 gal of water.

Based on comparisons of the RSSCT and pilot-column results for MWD (for both CRW and SPW) and the six participating utilities, it can be concluded that, for some waters, the RSSCT provides an economical means of estimating pilot- or full-scale breakthrough profiles that can be used in making preliminary assessments of carbon utilization rates and feasibility-level cost estimates. The limitations of the test, however, are not yet well defined, and for some waters (Philadelphia, for example) it appears that the RSSCT and pilot-scale breakthrough curves may not be comparable.

Cost estimates

Using the TOC breakthrough profiles and TOC/ $SDSTHM_{C1}$ relationships developed for both of Metropolitan's source waters—CRW and SPW—and the waters of the six participating utilities, cost estimates associated with implementing postfiltration GAC adsorption treatment were developed in relation to each of the $SDSTHM_{C1}$ treatment goals (i.e., 5, 10, 20, and 50 $\mu\text{g/L}$).

Methods. The cost equations used in calculating the GAC treatment cost estimates presented in this article are those developed by the USEPA from studies of field-scale systems.¹¹ More recent, modified versions¹² that reflect a reevaluation of the earlier equations in relation to newly obtained costs from completed field projects were incorporated into the final model. The equations were compiled in a spreadsheet cost model suitable for use on a personal computer. The cost equations include the most current USEPA estimates on all components of a GAC facility for water treatment. Details on these equations are contained in the cited references.

The two primary assumptions made in developing these costs involved the use of (1) gravity-flow concrete contactors for postfiltration GAC adsorption and (2) on-site carbon reactivation using fluidized bed furnaces. Other assumptions made for design and operating variables are listed in Table 5. As noted in the table, each of these values was determined from a different source. Those parameters specified by MWD will change when the equations are applied to other utilities and other locations. Material costs and cost indexes will also change in other locations.

Several steps were taken in developing the cost estimates for MWD. First, treatment parameters associated with the EBCTs used in the pilot study (i.e., 7.5, 15, 30, and 60 min) were determined under the premise of simultaneous THM precursor breakthrough for all columns and, therefore, regeneration of all contactors at the same time. Simultaneous breakthrough-reactivation is one of the assumptions incorporated into the USEPA cost model¹² and the GAC cost estimates.

Second, using one of two approaches—mathematical model simulation (using the plug-flow, pore and surface diffusion model) or graphical estimation based on pilot contactor breakthrough curves—calculations were made of regeneration frequencies for several intermediate EBCTs (i.e., EBCTs not tested in the pilot study) at the four SDSTHMC treatment goals and of treatment costs associated with each.

Third, total annual cost versus EBCT was plotted for each of the treatment goals. From this plot, the optimum EBCT associated with each treatment objective was determined.

Finally, for each of the optimum EBCTs determined in the preceding step, the costs of GAC treatment were reevaluated assuming effluent blending and staggered regeneration.

Using these steps, final cost estimates were developed for all of MWD's treatment plants. A comparison between cases of blended and unblended effluent

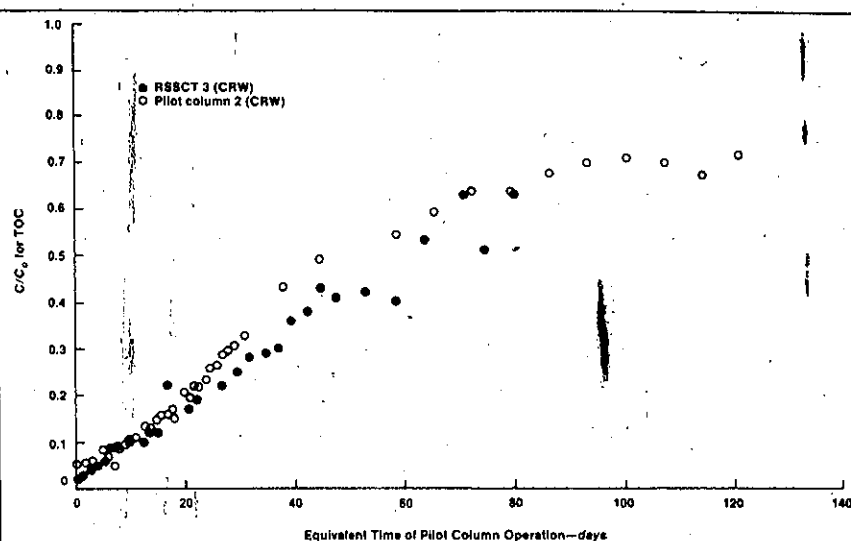


Figure 12. Comparison of TOC breakthrough profile from RSSCT with that from pilot GAC column (setup of RSSCT assumed intraparticle diffusivity to be a linear function of GAC particle size)

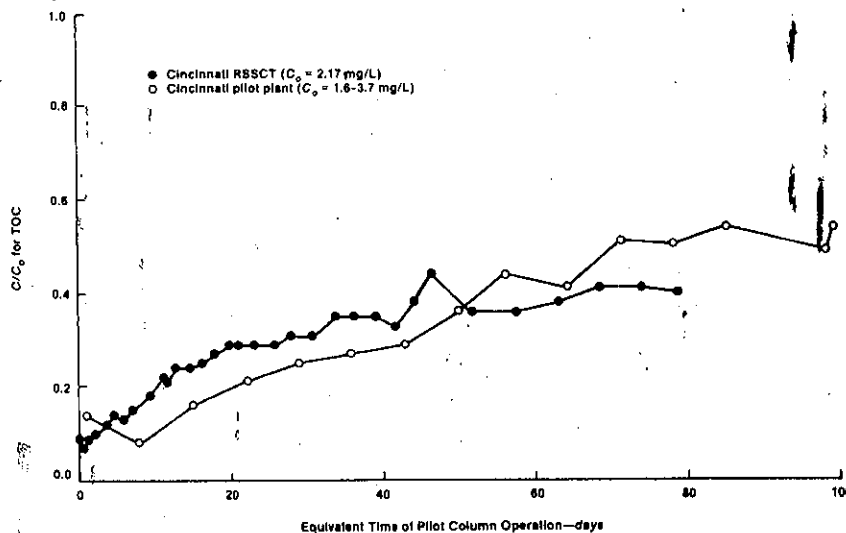


Figure 13. TOC breakthrough profiles for RSSCT and pilot plant for Cincinnati (EBCT—15 min)

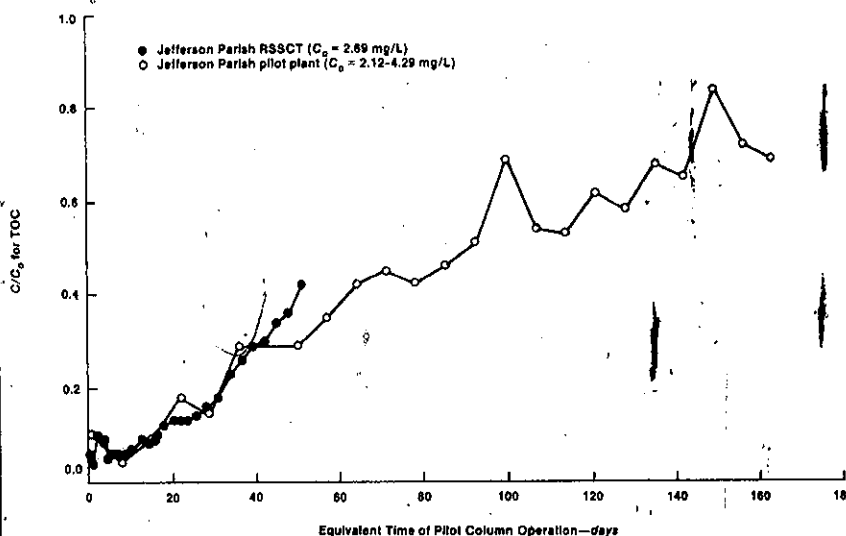


Figure 14. TOC breakthrough profiles for RSSCT and pilot plant for Jefferson Parish (RSSCT EBCT—15 min; pilot EBCT—20 min; pilot run 3)

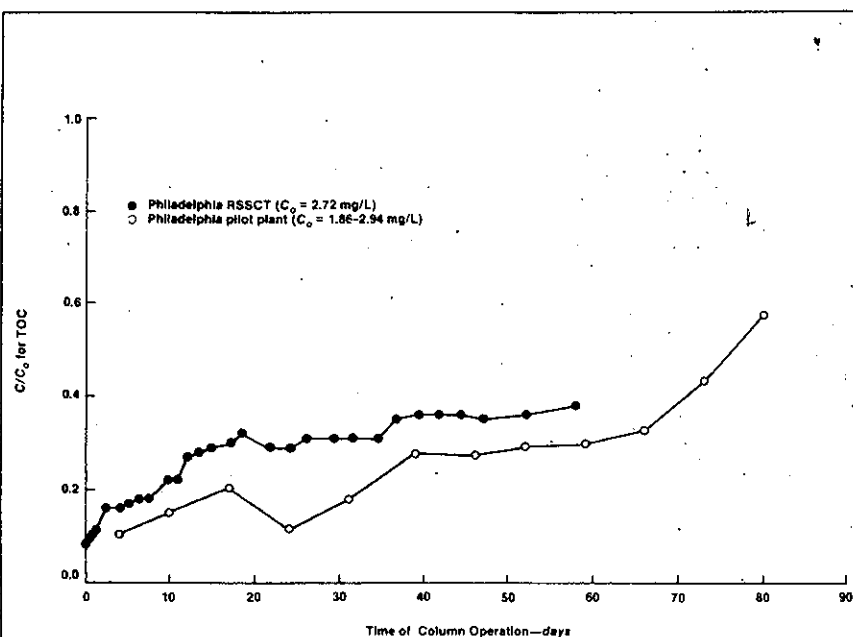


Figure 15. TOC breakthrough profiles for RSSCT and pilot plant for Philadelphia (RSSCT and pilot EBCTs—15 min)

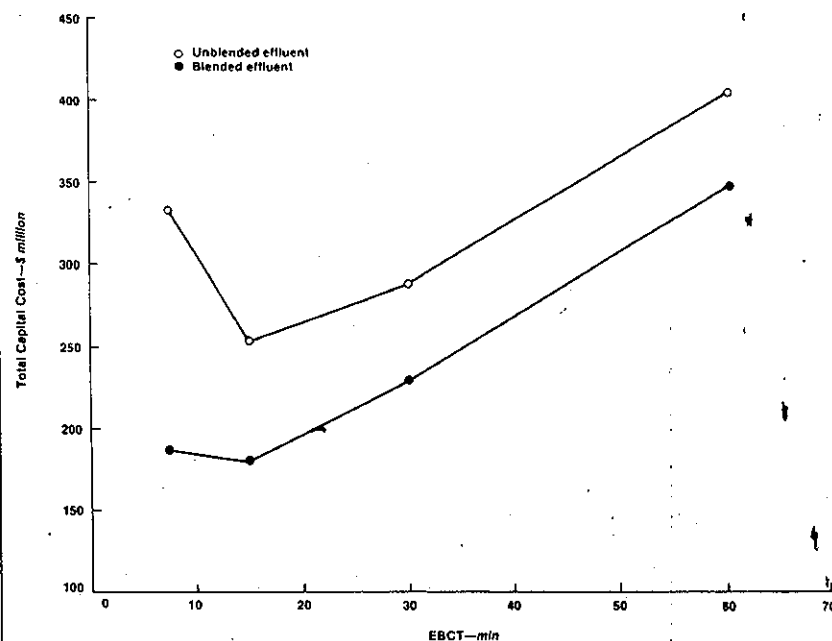


Figure 16. Total capital costs for blended and unblended effluents for Metropolitan treatment plant example (capacity—400 mgd; $SDSTH_{Cl}$ —20 $\mu\text{g/L}$)

for an example treatment plant with a capacity of 400 mgd is shown in Figure 16. As indicated, a substantial savings is realized for the blended effluent case.

The reason for using the case of a blended effluent is that it represents a more realistic scenario than assuming regeneration of all contactors at once. In actual operations, regeneration would

be on a staggered basis, as is filter backwashing in conventional plants. Because the effluent from all parallel contactors is blended prior to final disinfection, storage, and distribution, control of regeneration should be based on the TOC concentration of the blended plant effluent rather than on the effluent concentration from an individual con-

tactor. Under this assumption, the useful life of individual contactors is extended, thus reducing costs.

MWD. It has been proposed that MWD's treatment system capacity be expanded to 2,970 mgd by the year 2000. This capacity was used in preparing the GAC treatment cost estimates for MWD's treatment system as a whole. Cost estimates were also calculated for the individual water treatment facilities, as reported in the main body of the final report for this project.³ Assessment of the optimum EBCTs for the four $SDSTH_{Cl}$ treatment goals and the associated regeneration frequencies for blended effluent water are reported in Table 6.

The resultant treatment costs (i.e., capital, operation and maintenance, and annual unit costs) for MWD's total system are summarized in Table 7. The annual unit cost is the sum of the annual operations and maintenance cost and the annualized capital cost shown on the basis of a unit volume of water (e.g., 1,000 gal or 1 acre-ft).

Table 7 illustrates that GAC is an expensive means of THM control in MWD's system. In Southern California, a family of five uses about 1 acre-ft of water per year. MWD currently wholesales untreated water to its member agencies for \$197/acre-ft. A treatment surcharge of \$33 per acre-ft is added for wholesale treated water. Table 7 shows that the treatment surcharge would have to be increased by an amount ranging from 290 to 14,500 percent, depending on the target MCL for THMs. Overall, the annual cost of water to a typical family in Southern California would increase by 40 to 210 percent.

Participating utilities. A major objective of this study was to prepare cost estimates using RSSCT results, for the six participating utilities. Other researchers have discussed the limitations associated with scaling up small-scale column tests to full scale.¹³ Because only one RSSCT was performed for each utility, simulating a full-scale contactor EBCT of 15 min, the optimum EBCTs related to the different treatment goals could not be determined. In all cases, however, effluent blending was evaluated. Because of inadequate breakthrough curves for Atlanta and the East Bay Municipal Utility District, cost estimates could not be calculated for these two utilities. The capital and annual unit costs developed for the remaining four utilities are summarized in Table 8. The cost data in Table 8 also show that GAC would be an expensive means of meeting low THM standards. Palm Beach County, with its relatively high TOC levels, would be a particularly expensive place to control THMs with GAC.

These results confirm several of the findings published in a recent compila-

TABLE 5
Basic cost model input data

Criteria Category	Value
Filter bed depth*	10 ft
Filter surface area*	1,500 sq ft
GAC loss rate†	11 percent
GAC cost*	\$1.10/lb
GAC bulk density*	26 lb/cu ft
Fluid bed reactor capacity†	50,000 lb/d
Utility water rate‡	\$0.71/1,000 gal
Reactivation water use†	21 gal/lb GAC
Reactivator uptime‡	75 percent
On-site GAC transport labor†	0.4 h/1,000 lb GAC
On-site GAC transport water use†	2 gal/lb GAC
Electricity cost‡	\$0.075/kW-h
Labor cost‡	\$30/h
Diesel fuel cost*	\$0.90/gal
Construction cost index§	5,448
Producer's price index§	297.6
Engineering-administration‡	18 percent
Contractor's overhead and profit‡	12 percent
Contingencies‡	15 percent
Interest rate‡	8 percent
Amortization period‡	20 years
Cost adjustment factor (yard piping, 10%; low lift pumping, 8%; other site items, 8%; residuals handling, 4%)*	1.3
Average design capacity factor‡	54 percent

*Assumed design values

†Design-operating values specified by cost model developers

‡Parameters specified by MWD

§Published indexes

TABLE 6
EBCTs and GAC regeneration frequencies for different SDSTHM goals (blended effluent)—MWD system

SDSTHM _{C1} Goal µg/L	*Optimum EBCT min	GAC Regeneration Frequency days
5	60	42
10	30	47
20	15	32
50	7.5	30

TABLE 7
Cost estimates for total MWD system (total capacity 2,970 mgd)

Cost Category	Cost Estimate			
	5 µg SDSTHM/L (0.16 mg TOC/L)*	10 µg SDSTHM/L (0.25 mg TOC/L)	20 µg SDSTHM/L (0.44 mg TOC/L)	50 µg SDSTHM/L (1.0 mg TOC/L)
Capital—\$ million	3,900	1,980	1,300	730
O&M—\$ million/year	430	200	140	76
Annual unit—\$/1,000 gal†	1.46	0.68	0.47	0.29
Annual unit—\$/acre-ft	480	220	150	95

*The SDSTHM-TOC relationship is extrapolated from experimental data for TOC concentrations

>0.75 mg/L

†Includes annual amortized capital and O&M costs

TABLE 8
Summary of costs for participating utilities

Utility	Plant Capacity mgd	Capital Cost—\$ million				Annual Unit Cost—\$/1,000 gal*			
		5 µg SDSTHM/L	10 µg SDSTHM/L	20 µg SDSTHM/L	50 µg SDSTHM/L	5 µg SDSTHM/L	10 µg SDSTHM/L	20 µg SDSTHM/L	50 µg SDSTHM/L
Cincinnati	235†	689	149	74	‡	5.34	0.93	0.30	‡
Jefferson Parish	70	482	184	33	‡	13.40	4.82	0.52	‡
Palm Beach County	16	148	42	31	26	19.06	4.42	3.06	2.21
Philadelphia	320	634	152	‡	‡	3.38	0.58	‡	‡

*Includes annual amortized capital and O&M costs

†GAC capacity—175 mgd

‡No value reported because breakthrough was not reached during the RSSCT for this SDSTHM

tion of USEPA-funded GAC research that took place in the late 1970s and early 1980s.¹⁴ Although these studies were not designed to capture early TOC-SDSTH_{Cl} breakthrough—and their results were somewhat confounded by chlorination of the GAC filter influents—similarly high GAC treatment costs were determined for controlling THMs to levels of 10, 25, and 50 µg/L. Cincinnati was not representative of the other utilities, and the cost developed in those studies—\$0.149/1,000 gal to meet a THM standard of 25 µg/L—can be compared with a cost of \$0.30/1,000 gal to meet a 20-µg/L standard in this study.

Summary and conclusions

The principal conclusions that can be drawn from the data presented in this article are listed in the following sections.

TOC-SDSTH_{Cl} breakthrough curves. The CRW and SPW exhibited nonadsorbable TOC fractions of approximately 0.2 and 0.1 mg/L, respectively. These nonadsorbable TOC concentrations corresponded to SDSTH_{Cl} concentrations of approximately 5 µg/L for both waters. Because the lowest achievable THM concentration is fixed by the SDSTH_{Cl} potential of the nonadsorbable TOC, this in turn fixes the lowest THM level achievable. Allowing for some variability in influent conditions, the lowest allowable THM concentration in a GAC plant effluent would be about 10 µg/L.

The SDSTH_{Cl} technique was an excellent means of simulating THM formation in the distribution system.

TOC proved to be a suitable surrogate for SDSTH_{Cl} for both CRW and SPW. The relationship between SDSTH_{Cl} and TOC, however, was different for the two sources. Ultraviolet absorbance was an excellent surrogate for predicting TOC breakthrough.

Cost estimates. GAC adsorption is an expensive means of THM control. If MWD were to implement GAC adsorption as a unit process for THM precursor removal, the capital costs would range from \$730 to \$3,900 million (based on a capacity of 2,970 mgd), depending on whether the upcoming regulations are set at 50 or 5 µg/L.

Based on the RSSCTs performed for the participating utilities, the cost of implementing GAC adsorption would range from \$0.30 to \$3.06/1,000 gal for an SDSTH_{Cl} treatment goal of 20 µg/L. The cost would range from \$3.38 to \$19.06/1,000 gal for a goal of 5 µg/L. For an SDSTH_{Cl} treatment goal of 50 µg/L, cost estimates could only be developed for Palm Beach County, for which the implementation cost was estimated to be \$2.21/1,000 gal.

Optimizing the GAC cost model to include effluent blending is a much more realistic approach to developing cost

estimates than assuming simultaneous GAC regeneration for all columns.

DBPs. The THMs are the predominant DBPs formed during postdisinfection of GAC effluents with chlorine or chloramines. Control of the THM concentration by GAC should, at the same time, control the concentration of other DBPs in the distribution system. Breakthrough profiles for SDSTH_{Cl} appeared to track breakthrough of SDSTH_{Cl} and therefore support the use of THM control by GAC as an indication of the control of other DBPs.

Substantially higher SDSTH_{Cl} concentrations were formed during post-chlorination than were formed during postchloramination.

RSSCTs. The RSSCT was shown to be an economical means of estimating pilot-scale TOC breakthrough profiles, which can be used in making preliminary assessments of carbon utilization rates and feasibility-level cost estimates.

If the RSSCT is used in lieu of pilot-scale adsorption tests, the user should understand that the limitations of the test have not yet been clearly defined. However, two limitations are known:

- An RSSCT is performed over a short period of time; a pilot study requires much more time. Therefore, if the influent adsorbate concentration is not relatively constant, the RSSCT may not be able to produce a breakthrough profile that would be comparable with a pilot-scale profile.

- Selection of the appropriate scaling equations for sizing the RSSCT depends on the functional relationship between surface diffusivity and GAC particle size. Therefore, it is strongly recommended that this relationship be evaluated prior to performing the RSSCT.

If a pilot-scale evaluation is not an option, the RSSCT is a much more appropriate means of developing cost estimates than using either isotherm data or no adsorption data at all.

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CHAPTER 4

TRIHALOMETHANES IN U.S. DRINKING WATER—NORS TO ICR

Michael J. McGuire and Nicole Graziano

ABSTRACT

The National Organics Reconnaissance Survey (NORS) and the National Organics Monitoring Survey (NOMS), both conducted in the mid- to late-1970s, led to promulgation of the total trihalomethane (TTHM) regulation in 1979. In 1987, the American Water Works Research Foundation (AwwaRF) supported a survey in which more than 727 utilities submitted quarterly TTHM averages over a three-year period (1984–86). Detailed TTHM data were also collected as part of the Information Collection Rule (ICR) implemented from July 1997 through December 1998. This chapter presents a thorough analysis of TTHM occurrence in ICR systems and compares that occurrence with the findings from previous surveys.

Many groundwater and surface water TTHM samples collected under the ICR contained far higher TTHM concentrations than the 0.10 mg/L maximum contaminant level (MCL) in effect at the time. Apparently, the number of TTHM values exceeding the standard was so high because the MCL was based on running annual averages, in which high values can be averaged with lower concentrations to achieve compliance with the MCL.

The AwwaRF survey clearly showed a decrease in TTHM concentrations in water systems compared with the NORS and NOMS results, reflecting the implementation of TTHM control strategies by utilities. However, ICR results for 469 treatment plants and 10 blended distribution systems showed only a slight reduction in national TTHM occurrence since 1984–86. Although some utilities had installed advanced treatment processes in the 1990s in anticipation of promulgation of the Disinfectants/Disinfection By-products Rule, it appears that these process upgrades did not dramatically affect national TTHM concentrations at the time the ICR data were collected.

Nevertheless, a comparison of national TTHM occurrence surveys conducted from 1975 to 1998 illustrates a significant decline in TTHM concentrations during that period. Four descriptive statistics for NORS and ICR finished water data indicate that TTHM concentrations have been reduced by 50–60% over the 24-year period.

INTRODUCTION

In 1974, trihalomethanes (THMs) were discovered in treated drinking water around the world (Bellar, Lichtenberg, and Kroner 1974; Rook 1974; Rook 1976). To determine the national distribution of total THMs (TTHM) in the United States, the National Organics Reconnaissance Survey (NORS) was conducted in early 1975 (Symons et al. 1975) and was quickly followed by

the National Organics Monitoring Survey (NOMS), which confirmed the findings of NORS (Munch, Feige, and Brass 1977; Brass et al. 1977). On the basis of these two national surveys, the health effects data available at the time, and limited cost information, the U.S. Environmental Protection Agency (USEPA) in 1979 established the TTHM maximum contaminant level (MCL) at 0.10 mg/L (USEPA 1979).

Subsequently, one minor and two major national TTHM surveys were conducted to determine the distribution of TTHM in U.S. drinking water. The most comprehensive survey of TTHM data occurred as part of the Information Collection Rule (ICR), which gathered data in 1997 and 1998 (USEPA 1996).

This chapter summarizes and analyzes the TTHM data collected as part of the ICR and compares them with TTHM data collected as part of several other national surveys.

NATIONAL SURVEYS TO ESTIMATE TTHM OCCURRENCE

Several estimates of TTHM occurrence in U.S. drinking water were made over the 24-year period 1975–1998. NORS was designed to identify national TTHM occurrence. Samples were collected from finished water locations at individual plants of the subject utilities and shipped back to the USEPA laboratory in Cincinnati for analysis. After each sample was collected and prior to shipping, a dechlorinating agent was added to the sample bottle to stop further THM production, and the sample was iced to preserve TTHM concentrations.

As a follow-up to the initial USEPA survey, a second survey, called NOMS, was designed to increase understanding of THM formation kinetics and to determine the best method of preserving samples collected as part of a survey. NOMS was actually a series of three surveys in which sample bottles were sent to 113 U.S. cities in 1975–76 (Munch, Feige, and Brass 1977; Brass et al. 1977). Samples collected under Phase 1 of NOMS were dechlorinated upon collection, making these results most comparable to the NORS data. All samples were sent to the USEPA laboratory in Cincinnati for analysis.

On January 14, 1987, 1,255 survey questionnaires were sent to water utilities serving more than 10,000 people as part of an investigation supported by the American Water Works Association Research Foundation (AwwaRF survey). The questionnaires asked each utility to compile quarterly average TTHM data over the period 1984–1986 and send them to a central

database. The survey population consisted of all utilities serving more than 50,000 people and a sampling of utilities serving 10,000 to 50,000 people. A significant number of utilities (727 out of 1,255) submitted TTHM data (McGuire and Meadow 1988).

Four quarters of data on disinfection by-products (DBPs) were collected from 35 utilities (mostly large and very large utilities) from spring 1988 to winter 1989 (35-city survey). Samples for this survey were collected from clearwell effluents and were dechlorinated before being shipped to a central laboratory at 4°C. These data do not represent the maximum DBP concentrations that can occur in a distribution system (Krasner et al. 1989). Results from the 35-city and AwwaRF surveys were used in regulatory negotiations in 1992–93 and in the Federal Advisory Committee Act (FACA) negotiating process in 1997; they were instrumental in the development of the Disinfectants/DBP Rule, which was promulgated December 16, 1998, and became effective January 1, 2002 (Roberson et al. 1985, USEPA 1998).

As part of the ICR monitoring program, six quarters of DBP data were collected from the distribution systems of 296 utilities (500 treatment plants) during the period July 1997 to December 1998 (USEPA 1996). All U.S. utilities serving populations in excess of 100,000 people participated in ICR monitoring. Thus, the TTHM data from the ICR survey represents a census of TTHM data from very large utilities. The ICR was the most complex and expensive data-gathering effort from drinking water utilities ever attempted in the United States. ICR samples produced DBP data from five sampling points within each participating utility: the finished water (FIN) sampling point, a distribution system equivalent (DSE) location (a distribution site with well-characterized detention time), two locations representing average distribution system detention times (AVG1 and AVG2), and the location representing maximum distribution system detention time (MAX). USEPA-approved laboratories analyzed all ICR DBP samples.

METHODS

For the analyses presented in this chapter, ICR TTHM data were extracted from the ICR Auxiliary 1 database, version 5.0, which contains data from 296 utilities and DBP information from 500 water treatment plants. TTHM values that were below minimum reporting levels were replaced with zero values before further calculation or graphing. Although the ICR database

contains analytical results from many sampling locations, this chapter focuses on results from the finished water sampling location and the four distribution system sampling points.

In several cases, the number of data points from which statistics are derived is different because of missing data. For example, if data from one of the four THMs was missing, no TTHM value was calculated. Null TTHM values were not included in any averages or graphed on any figures. Also, because water wholesalers did not collect samples from any distribution system, their treated water TTHM data are found only in the statistics on FIN locations. In addition, ICR sampling included 10 utilities that distributed blended water, and the distribution system data for these utilities are not directly related to a single treatment plant.

The descriptive statistics summarized in the tables in this chapter are consistent with common usage. Mean refers to the arithmetic average of all data. Median represents the 50th percentile of the data, with half the data points above the median and half below. P75 and P90 refer to the 75th and 90th percentile values that describe the TTHM concentrations for which 75% and 90% of the data are less than the listed numbers.

ANALYSIS OF ICR TTHM DATA

The characteristics of THM formation have been studied ever since the discovery of these DBPs in 1974. Factors influencing the amount and species of THM produced have been evaluated in numerous laboratory studies, including studies on the amount and type of organic precursors (Stevens et al. 1976) and studies on the influence of bromide concentration, water temperature, and amount and type of disinfectant (Symons et al. 1982). Using ICR TTHM data, this section examines these factors and others on a nationwide basis.

Table 4.1 summarizes all ICR TTHM data collected from all utilities that reported this information (including blended distribution systems). A total of 30,291 TTHM data points were used to construct this table.

Table 4.1
Summary of ICR TTHM results

TTHM parameter	Measure	Finished water	Distribution system sampling sites				All distribution system sites
			DSE	AVG1	AVG2	MAX	
CHCl ₃	Count	2,121	1,873	1,858	1,851	1,935	7,517
	Mean†	16.7	21.7	22.9	23.3	25.8	23.5
	Median†	12.0	16.0	17.9	17.3	19.3	17.7
	P90†	39.6	50.9	50.0	51.9	59.9	53.0
BDCM	Count	2,139	1,891	1,879	1,873	1,960	7,603
	Mean	6.7	8.1	8.4	8.4	8.9	8.4
	Median	5.0	6.5	6.8	6.7	7.3	6.8
	P90	16.0	18.0	19.0	19.0	19.2	19.0
DBCM	Count	2,136	1,887	1,877	1,871	1,953	7,588
	Mean	3.4	4.0	4.3	4.2	4.5	4.3
	Median	1.4	1.9	2.0	2.0	2.2	2.0
	P90	10.0	11.0	12.0	11.5	12.3	12.0
CHBr ₃	Count	2,135	1,887	1,873	1,868	1,955	7,583
	Mean	1.0	1.2	1.4	1.4	1.5	1.4
	Median	0.0	0.0	0.0	0.0	0.0	0.0
	P90	2.9	3.4	3.9	3.7	4.3	3.8
TTHM	Count	2,108	1,865	1,852	1,843	1,921	7,481
	Mean	28.0	35.0	37.0	37.3	40.8	37.6
	Median	23.3	30.4	32.5	33.2	35.8	33.0
	P90	60.2	73.8	75.1	77.5	84.7	78.3

* Statistics include TTHM data for blended distribution systems.

† All mean, median, and P90 values are in µ/L.

As other studies have noted, chloroform (CHCl₃) dominated the three other THM species and was found at the highest mean concentration, 23 µg/L. Mean concentrations of bromodichloromethane (BDCM) at 8 µg/L and dibromochloromethane (DBCM) at 4 µg/L were far lower than the mean concentration of CHCl₃, and the overall mean concentration of bromoform (CHBr₃) was 1 µg/L. Overall mean and median values for the entire ICR TTHM data set were 38 and 33 µg/L, respectively.

Figure 4.1 presents box-and-whisker plots of the concentrations of the TTHM component species listed in Table 4.1. Figure 4.1 was constructed with all six quarters of TTHM data using the graphing program Statistica. As indicated in the figure legend, this box-and-whisker plot describes the 10%–90% range of concentrations, designated by the whiskers, and the interquartile range (25%–75%), illustrated by the boxes. Data points that differ substantially

from the central tendency of these data sets are identified as outliers and extremes. In this chapter, outliers are defined as values greater than 1.5 times the interquartile range; extremes are defined as values more than 3.0 times the interquartile range. Plots for the bromine-substituted THM species (BDCM, DBCM and CHBr_3) indicate significant distributions of outliers and extremes, but these are nothing compared with the wide range of outliers and extremes shown above the box-and-whisker plot for CHCl_3 .

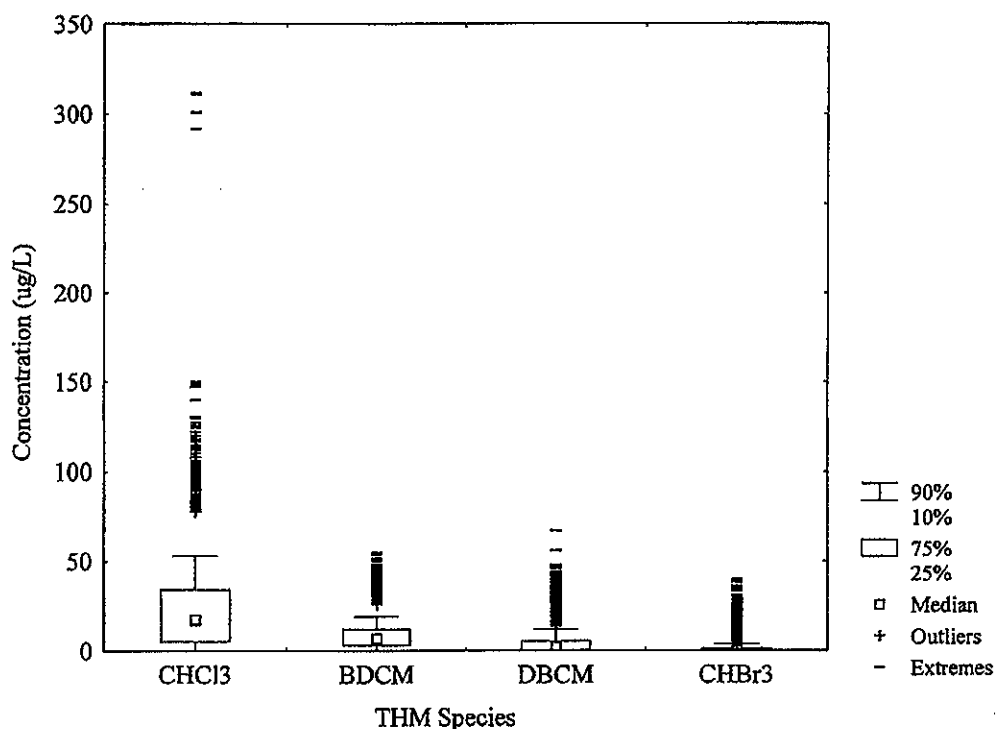


Figure 4.1 Comparison of ICR results for individual TTHM species, showing outlier and extreme values, over six quarters (TTHM concentrations in purchased and mixed supplies were included; those in finished water were excluded)

TTHM Variability by Source of Supply

Chapter 1 of this book describes how the influents of ICR treatment plants were characterized with respect to the predominant source of supply (Wysock et al. 2002). The influent of most plants consisted of surface water or groundwater supplies. Figure 4.2 compares TTHM data from the distribution systems of all ICR utilities with TTHM data from the distribution systems of ICR utilities treating surface water or groundwater. In Figure 4.2, the four TTHM values above 300 $\mu\text{g/L}$ were from the distribution system of a surface water utility in Mississippi. In Figure 4.1, these extreme values were shown to be composed primarily of CHCl_3 . Figure 4.2 also shows that the total number of TTHM samples from all six ICR quarters is 760 more than the sum of all TTHM samples from plants treating surface water and ground water. The differential is due to the inclusion of TTHM data from blended distribution systems in the "ALL" category.

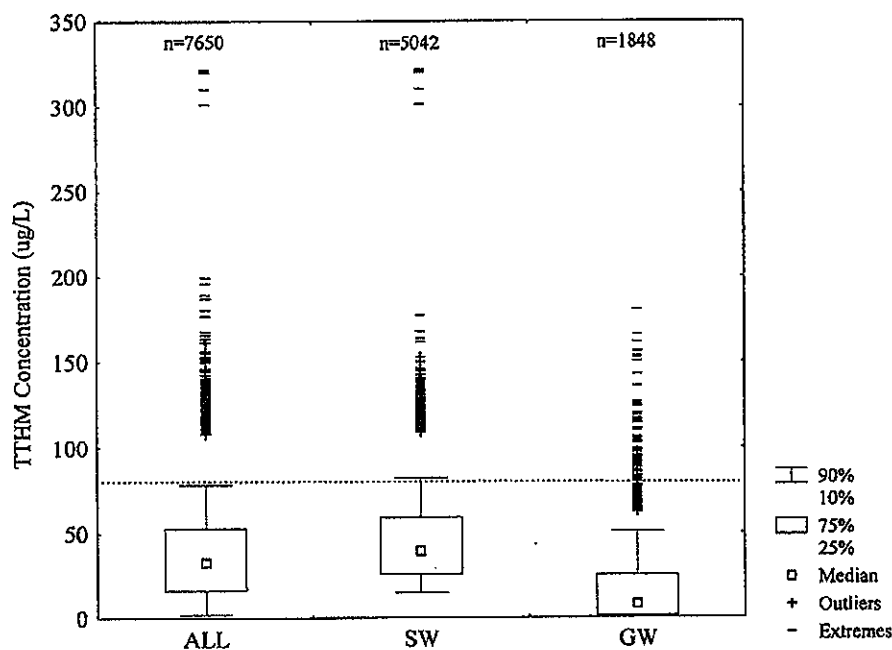


Figure 4.2 Comparison of TTHM results from the distribution systems of ICR surface water (SW) and groundwater (GW) utilities with TTHM results from all ICR distribution system data, showing outlier and extreme values, over six quarters

The distribution system TTHM data from plants using groundwater as a source of supply show a pattern similar to that of the data from plants using surface water (Figure 4.2). The median TTHM concentration in samples from groundwater systems is far lower (7.8 µg/L) than the median concentration in samples from surface water systems (39.3 µg/L). In general, the distribution systems of U.S. utilities that produce disinfected groundwater have very low TTHM concentrations, primarily as a result of very low concentrations of organic precursors in groundwater supplies. However, the TTHM data for ICR groundwater systems include outliers and extremes at significantly high concentrations.

Figure 4.2 shows that there are many exceptions to the low TTHM concentrations usually found in groundwater. Groundwater supplies in the southeastern United States are generally high in total organic carbon (TOC) (see Chapter 12, McGuire and Hotelling 2002). During presentations of these data to the FACA committee as part of regulatory negotiations, these outlier values for groundwater DBPs were called the "Florida effect." The data shown in Figure 4.2 confirm this effect. Of the 116 outlier and extreme values shown on the groundwater box-and-whisker plot, 68 were from groundwater utilities in Florida. The next most frequently listed state contributing high TTHM values from groundwater was New Jersey, which accounted for only 10 of the 116 outliers and extremes.

TTHM Variations Over Time

Figure 4.3, one of the graphs presented to the FACA committee in late 1999, shows quarterly median TTHM data for groundwater and surface water plants by distribution system sampling location. Plots of the surface water data show progressively higher median concentrations of TTHM as samples were collected from the finished water, DSE, average, and maximum sampling locations. Quarters 1 and 5, which comprised the summer months (July–September) during which ICR samples were collected, showed the highest median values for all sampling locations from surface water systems. Quarter 3, which represented the only winter months of ICR monitoring, showed the lowest median values. Median values for Quarters 2 and 6 (fall quarters) were virtually indistinguishable from one another. Median values for the spring quarter (Quarter 4) were only slightly lower than those for the summer quarters and were higher than those for the winter and fall quarters, possibly reflecting the impact of increased runoff of

organic precursors on TTHM production. Data for the groundwater systems show that none of the six quarterly median TTHM values changed during the 18-month ICR monitoring period because of the consistency (and low concentrations) of organic precursors in groundwater sources.

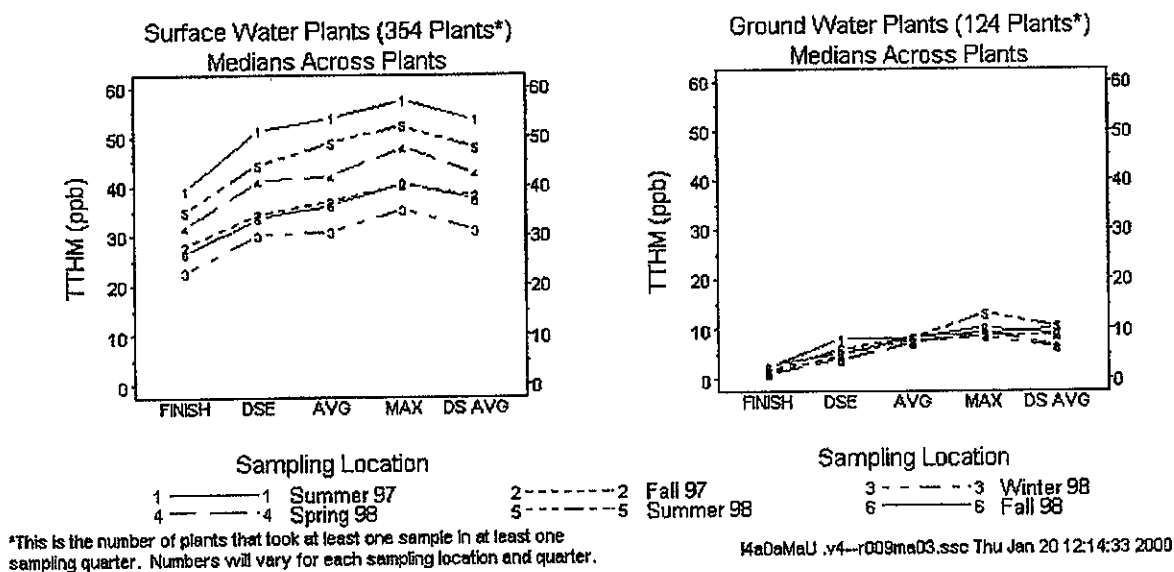


Figure 4.3 ICR TTHM data collected over six quarters and presented to the FACA Committee
(Source: Cadmus 2001)

Geographic Variability in TTHM Occurrence

Table 4.2 lists descriptive statistics for TTHM data associated with the 10 USEPA regions (Figure 3.2, Roberson 2002). TTHM data are not shown by state because the counts of plants and blended distribution systems by state are too low to allow meaningful statistical comparisons using quarterly TTHM data. The data in Table 4.2 indicate large differences in TTHM occurrence among USEPA regions. Mean TTHM concentrations ranged from 15 $\mu\text{g/L}$ in the Pacific Northwest and Alaska (Region 10) to 47 $\mu\text{g/L}$ in the Mid-Atlantic states of Virginia to Pennsylvania (Region 3). Median and P90 values showed the same trends—lowest to highest TTHM concentrations by region. Somewhat surprisingly, Region 3 had higher TTHM concentrations than Region 4, which includes southeastern states (including Florida) known to have the highest concentrations of organic precursors in their sources of supply. It is believed that utilities in Region 4 have already had to install treatment systems to reduce TTHM production in order to comply with the 1979 TTHM MCL of 0.10 mg/L.

Figures 4.4 and 4.5 plot the mean and P90 values from Table 4.2 on maps of the United States. Not surprisingly, Regions 3 and 4 show up in the highest category of mean TTHM concentrations (43–47 $\mu\text{g/L}$). Region 2 (New York, New Jersey, and Puerto Rico) has a high mean and a very high P90 TTHM value because of high organic precursor concentrations in the source water of the six ICR utilities located in Puerto Rico.

Table 4.2
ICR TTHM statistics* for 10 USEPA regions

USEPA region	Number of ICR plants	Mean ($\mu\text{g/L}$)	Median ($\mu\text{g/L}$)	P90 ($\mu\text{g/L}$)	Count
1	16	44.2	42.0	74.3	298
2	50	39.9	34.1	86.0	694
3	39	47.1	42.4	86.7	624
4	97	42.8	37.1	79.9	1,538
5	59	29.1	21.2	67.5	962
6	55	39.7	34.5	79.7	963
7	19	34.1	29.8	70.0	446
8	18	32.6	29.1	52.9	317
9	104	35.6	33.2	79.9	1,541
10	14	15.2	10.5	36.7	254
Total: 471					

* Statistics exclude finished water concentrations but include values from blended systems.

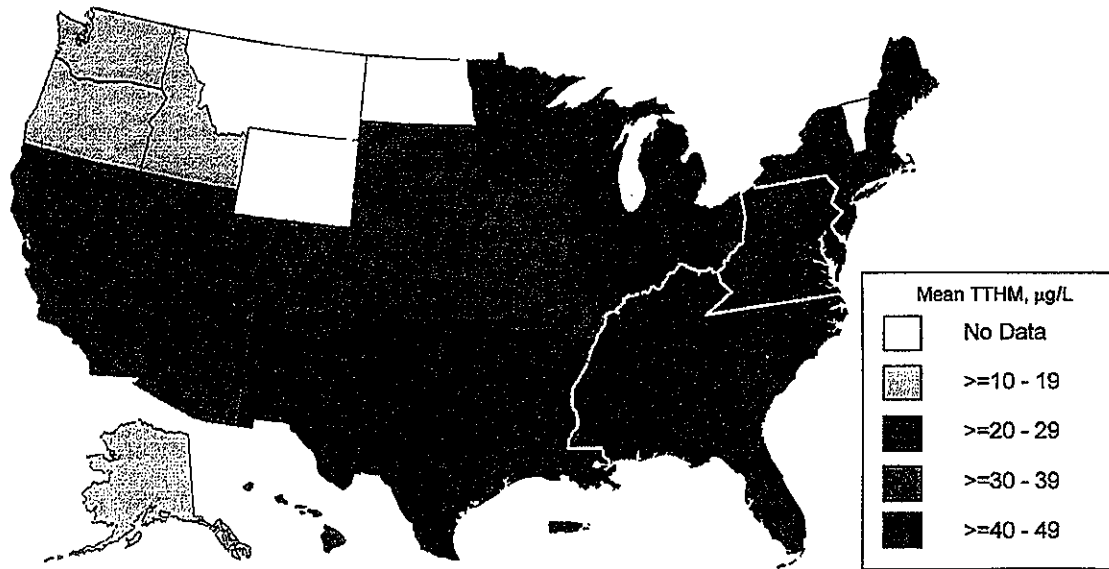


Figure 4.4 Mean TTHM concentrations by USEPA region—running annual averages for six quarters

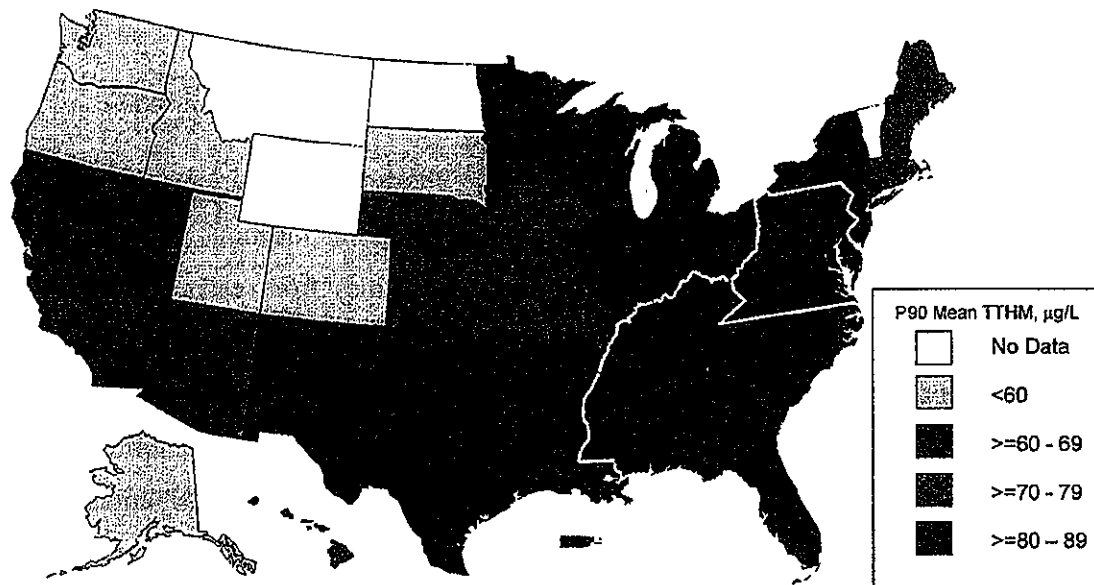


Figure 4.5 Ninetieth percentile mean TTHM concentrations by USEPA region—running annual averages for six quarters

It is somewhat surprising to see Region 1 (New England) in the category with the highest mean TTHM occurrence, given the long winters and cold water temperatures in this region. Inspection of the information on source water quality, TTHM concentrations, and treatment at the ICR utilities in this region shows that source water quality does not explain the high TTHM concentrations in the New England states. Instead, the explanation is that 15 out of 16 plants used free chlorine as both the primary and secondary disinfectant, a practice that can be expected to produce significant TTHM concentrations. (The exception is Portland, Maine, the only ICR utility in the region that used ozone and chloramines.)

Comparing Figures 4.4 and 4.5 shows that the highest categories of mean and P90 values stretch from Region 6 (Texas, Louisiana, Arkansas, Oklahoma, and New Mexico) to the regions dominated by East Coast states. High TTHM concentrations in these regions stem from a combination of high source water precursor concentrations, high temperatures, and, for many utilities, continued reliance on free chlorine for primary and secondary disinfection. The Great Lakes states (Region 5) have the next highest TTHM concentrations, followed by the western and midwestern states, which have moderate TTHM concentrations. The Pacific Northwest and Alaska (Region 10) enjoy the lowest TTHM concentrations in the country.

TTHM Variation Within Distribution System Locations

Figure 4.6 presents box-and-whisker plots for the first four quarters of ICR TTHM data from finished water, DSE, average, and maximum sampling locations plus the distribution system average (DS AVG) for each quarterly set of values from each plant. The data are presented separately for plants using surface water and groundwater supplies. There are clear differences between the data sets of surface water and groundwater systems, but within each data set, there are few differences in mean and median values from the various sampling locations.

The ICR TTHM data were based on treatment plants, not on distribution systems. For example, a utility might have three treatment plants serving its distribution system. Two of these plants might produce high TTHM concentrations, and one might use a groundwater source that forms low TTHM concentrations. Under the 1979 TTHM regulation, quarterly averages for such a utility are calculated not by averaging data from individual plants but by averaging all of the

data from all of the distribution system sampling locations over one quarter and then compiling a running annual average (RAA) of that data over four quarters.

Figures 4.3 and 4.6 are published on a Web site created by a company called SAIC and maintained for USEPA by the Cadmus Corporation (Cadmus 2001). For surface water supplies, the graphed data in Figure 4.6 show that overall TTHM average and median values for all the sampling points were less than 50 µg/L. All of the plants and utilities represented by the data in Figure 4.6 complied with the TTHM regulation. However, many individual data points and quarterly averages exceeded the 0.10 mg/L TTHM MCL in effect at the time of ICR sampling. A number of TTHM data points exceeded 200 µg/L, and four were well in excess of 300 µg/L.

On October 27, 1999, Figure 4.6 was presented to the FACA regulatory negotiation committee as part of a summary of ICR DBP data. Because of the high concentrations of the outliers and extremes shown on its plots, this figure had a greater impact on the committee than any other of the several hundred DBP graphs and tables presented over a nine-month period.

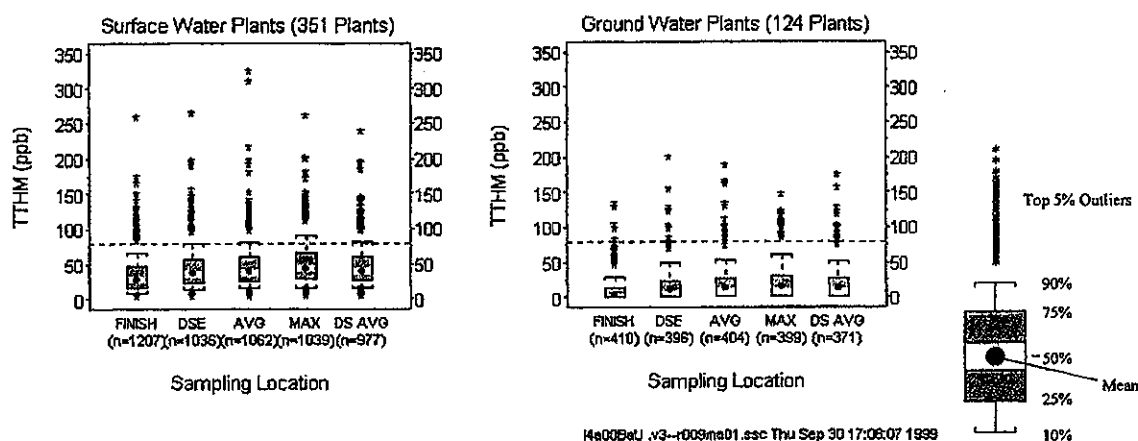


Figure 4.6 TTHM concentrations in the distribution systems of plants treating surface water and groundwater supplies (Source: Cadmus 2001)

FACA committee members were surprised and concerned that systemwide averaging under the 1979 TTHM regulation allowed what appeared to be significant TTHM concentrations in systems complying with the MCL. In an attempt to reduce the high concentrations of TTHM and other DBPs served to water utility customers, the FACA committee recommended that the USEPA incorporate the concept of locational running annual averages (LRAAs) in the Stage 2 DBP Rule to ensure compliance with the MCLs for TTHM and HAA5 (the sum of five haloacetic acids) (USEPA 2000). An LRAA is calculated by averaging TTHM values for each distribution system sampling location over four quarters. Use of an LRAA for compliance purposes eliminates the possibility of averaging high and low TTHM values throughout a distribution system over the same quarter.

Table 4.3 compares descriptive statistics for LRAA data with those for RAA data collected over all six quarters of ICR sampling. LRAA statistics show increasing TTHM concentrations as sampling progressed from finished water to MAX locations. The last two columns of Table 4.3 compare the highest LRAA mean, median, and P90 values for all of the locations with the highest mean, median, and P90 values for the RAA data points collected during ICR monitoring. The highest LRAA values were greater than the highest RAA values.

The last two columns in Table 4.3 suggest that meeting a TTHM MCL based on LRAA calculations would be more difficult than meeting an MCL based on RAA calculations. This finding is further strengthened by the fact that ICR RAA data were based on average values from only one treatment plant, whereas RAA calculations for MCL compliance have been based on averages of many distribution system samples possibly representing different sources of supply.

Table 4.3
Highest TTHM LRAA and RAA concentrations

TTHM	Highest LRAA values by sampling site†					Highest LRAA	Highest RAA
	FIN	DSE	AVG1	AVG2	MAX		
Count	481	434	433	433	441	496	448
Mean*	29.7	38.4	40.8	40.5	44.8	47.2	40.7
Median*	26.3	35.6	37.0	38.0	42.2	44.8	38.4
P90*	60.6	72.2	75.5	75.5	85.9	87.4	74.0

* All mean, median, and P90 values are in µg/L.

† Values for blended systems are included. LRAA values shown for each sampling location are those reported as the highest from the three annual periods comprising the 18-month IRC monitoring period.

The Impact of Treatment on TTHM Formation

It is useful to examine TTHM concentrations in the distribution systems of a variety of treatment plants to see if the type of treatment has a major effect on DBP production. Figure 4.7 presents a box-and-whisker plot of TTHM occurrence data based on treatment plant type, as defined in Chapter 3 of this book (Roberson 2002). Median values for all treatment plant categories shown on the graph were very close, except for those in the two groundwater treatment categories—DIS/GW and OTHER/GW—whose median values were lower than the others. The outliers and extremes in the groundwater treatment categories are interesting because most Florida plants were classified as softening plants. A number of the groundwater treatment facilities treating sources high in organic precursors were located outside the state of Florida.

Except in the groundwater treatment categories, the interquartile ranges in all of the treatment categories overlap, indicating little difference among them.

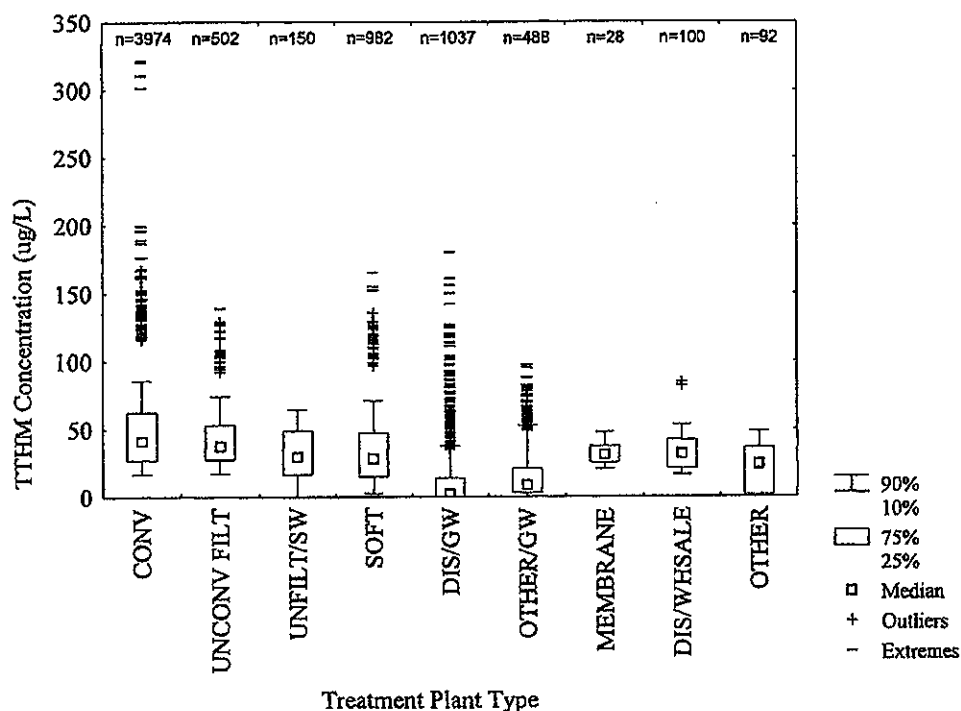


Figure 4.7 Comparison of TTHM concentrations in distribution systems as a function of treatment plant type (finished water samples were excluded; direct filtration, in-line filtration, and slow sand filtration plants were combined into the category for unconventional filtration; five types of softening plants were combined to form the category for softening plants)

The influence of disinfectant type is examined in Figures 4.8 and 4.9. All the data in Figure 4.8 are separated according to categories designating the primary disinfectant used at the plant: free chlorine at CL2 plants, some form of chloramine at CLM plants, free chlorine followed by chloramine at CL2_CLM plants, chlorine dioxide at CLX plants, and ozone at O3 plants. There is not much difference among the box-and-whisker plots for all of the disinfectant categories. Outliers and extremes appear to dominate the CL2, CLX, and O3 categories, but these data must be interpreted with care. First, the number of plants in the three categories varied widely; several dozen plants used chlorine dioxide and ozone, whereas hundreds used free chlorine. Also, plants that had experienced problems with high TTHM concentrations in the past and that had source water supplies with high concentrations of organic precursors had already changed to alternative disinfectants such as chlorine dioxide or ozone before ICR monitoring began. Even with the alternative disinfectants, however, outliers and extremes were produced. The four extreme TTHM concentrations from the plant in Mississippi were produced with two different disinfectants.

Figure 4.9 indicates that the type of disinfectant used in the distribution system is not likely to predict distribution system TTHM concentrations. For example, given equal source water TOC concentrations, using chloramine as the distribution system disinfectant would be expected to result in lower TTHM concentrations overall. However, plants using chloramine had most likely changed to this disinfectant because high organic precursor concentrations in their source water had made it difficult for them to meet the 1979 TTHM MCL. In other words, the TTHM concentrations had already been "adjusted" as a result of regulatory compliance efforts. Both free chlorine and chloramine systems had a number of outliers and extremes. Surprisingly, the four extremely high TTHM values (greater than 300 µg/L) occurred in a chloraminated distribution system, illustrating that converting to an alternative disinfectant is not a panacea.

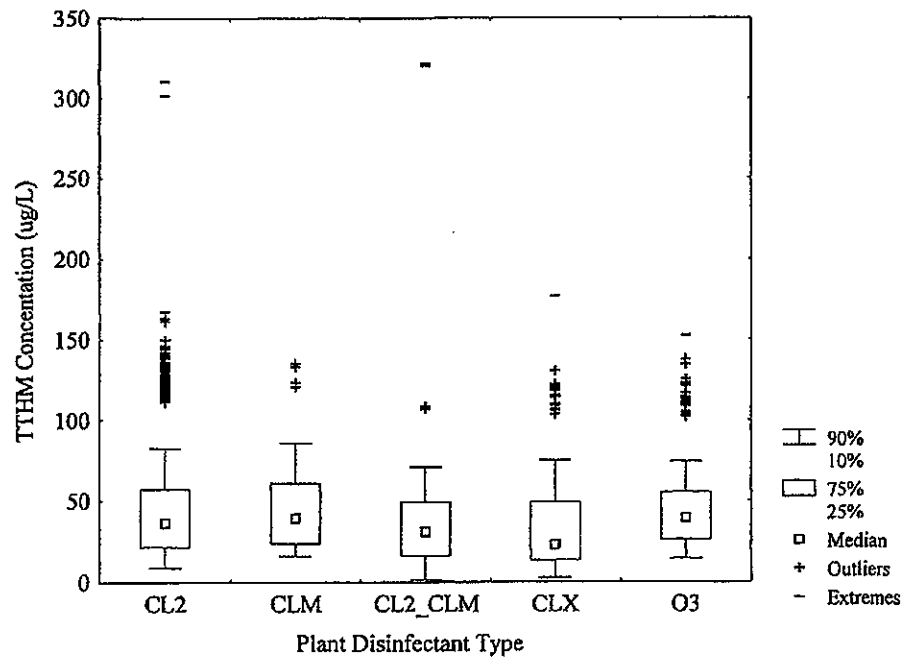


Figure 4.8 Comparison of distribution system TTHM concentrations as a function of type of disinfectant used in the treatment plant

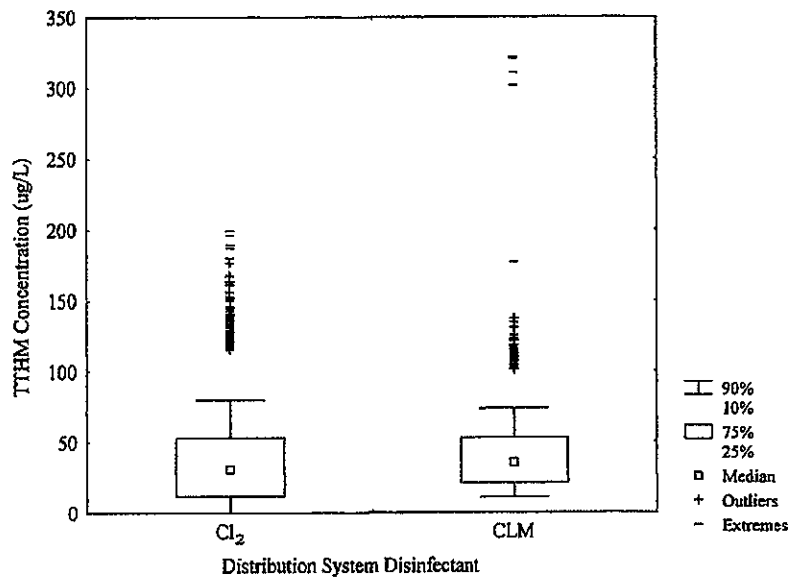


Figure 4.9 Comparison of TTHM concentrations as a function of type of disinfectant used in the distribution system

Influence of Source Water Quality on TTHM Formation

Figure 4.10 shows that TOC concentration in treatment plant influent is a poor predictor of TTHM concentration in the distribution system. TTHM values increase slightly with increasing TOC concentrations, but the data vary substantially. Median and P90 values both show a slight increase across the five categories of TOC concentrations (<1mg/L to >4 mg/L), but the box-and-whisker plots overlap in all five TOC ranges. Figure 4.11 illustrates the poor correlation between paired data sets of influent TOC and finished water TTHM concentrations. The coefficient of determination (r^2) value of 0.1336 confirms that there is virtually no relationship between these two parameters in these paired data sets.

Many laboratory studies have demonstrated the influence of TOC concentration on the amount of TTHM formed. An important graph (based on NORS data) in a USEPA publication summarizing TTHM control (their Figure 1) showed, roughly, that increasing TOC concentrations in source water resulted in higher TTHM concentrations in the effluents of NORS treatment plants (Symons et al. 1982). It is not surprising that a relationship between TOC and TTHM existed in 1975 because, at that time, utilities had not yet reduced chlorine dosages or installed TTHM control technologies. But the ICR data set clearly shows that influent TOC alone is a poor predictor of TTHM occurrence in a wide range of U.S. treatment plants. In individual treatment plants and in narrow ranges of influent water quality conditions, higher influent TOC concentrations can certainly cause higher TTHM concentrations in treated water. In fact, a variety of water treatment parameters and environmental conditions affect TTHM formation, and these factors are discussed in detail in Chapter 8 of this book (Swanson et al. 2002). TOC was not a good overall predictor of TTHM formation in the ICR data set because by the time these data were collected, utilities had adjusted their treatment processes and disinfectants to achieve lower TTHM concentrations in order to meet current and future regulations.

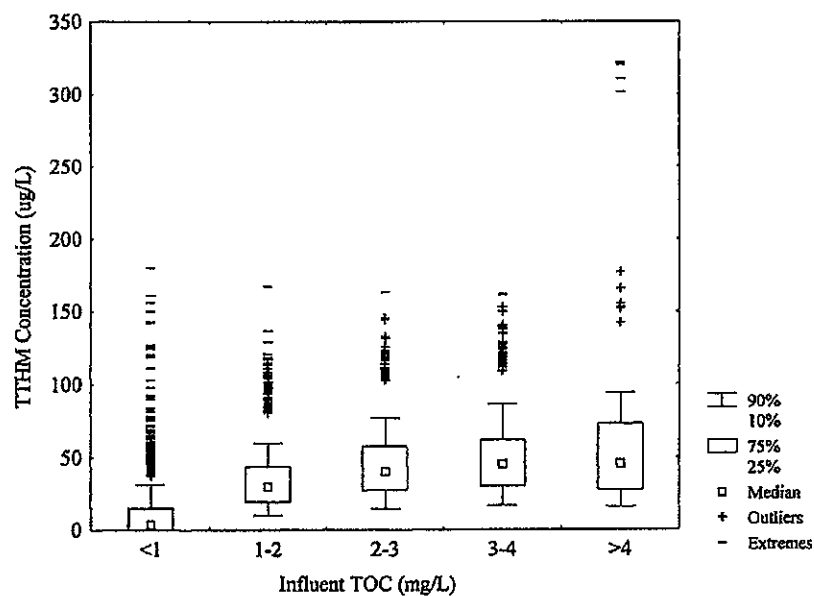


Figure 4.10 Comparison of mean distribution system TTHM concentration as a function of influent TOC concentration

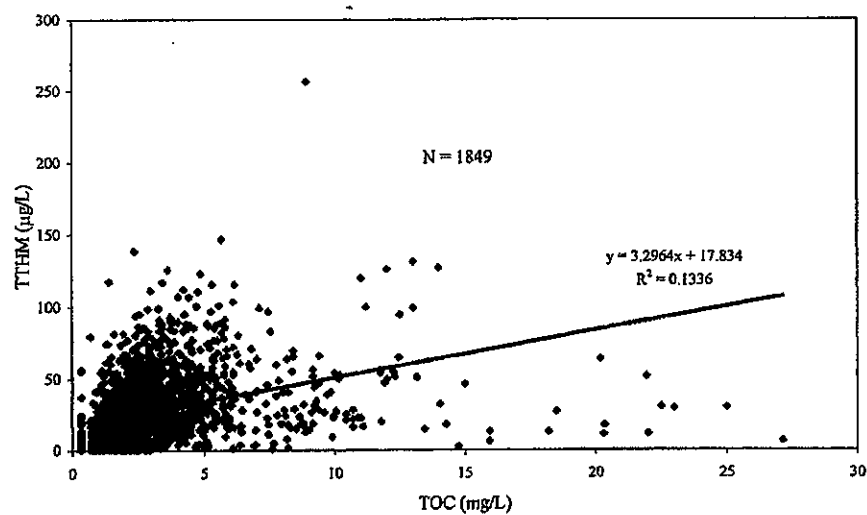


Figure 4.11 Influent TOC concentrations versus finished water TTHM concentrations in ICR plants

In laboratory studies, bromide ion concentrations have been shown to affect TTHM production. However, the bromide data summarized in Figure 4.12 show that TTHM concentrations in the distribution systems of ICR utilities were not well predicted solely by bromide ion concentrations in plant influent. For this data set, on a microgram-per-liter basis, increasingly high bromide concentrations appeared not to affect the TTHM values produced. Utilities with very high concentrations of bromine-substituted THMs had probably taken action to control DBPs by the time these data were collected, and such control strategies undoubtedly confounded the relationship between these two parameters. Chapter 6 (McLain, Obolensky, and Shukairy 2002) demonstrates that higher bromide ion concentrations led to a higher degree of bromine substitution in bromine-substituted DBPs measured under the ICR.

Figure 4.13 presents a three dimensional picture of the relationships between TOC and bromide concentrations in plant influent and mean TTHM concentrations in distribution systems. By factoring out the influence of bromide, Figure 4.13 shows a clearer relationship between TOC concentrations in plant influent and TTHM production. When bromide concentrations in plant influent were low or moderate, mean TTHM concentrations increased with increasing TOC concentrations in the source water. At the highest bromide concentration, TTHM appears to increase and then decrease with increasing TOC. Once again, it appears that plants whose source water poses the greatest water quality challenges (TOC >4 mg/L, bromide >100 μ /L, $N = 372$) have taken steps to control TTHM concentrations (see Chapter 12, McGuire and Hotelling 2002).

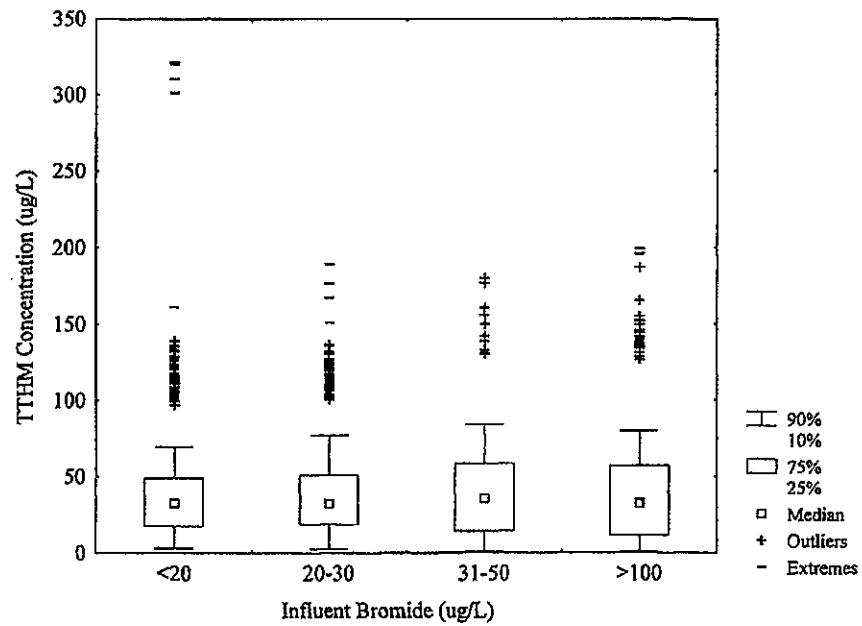


Figure 4.12 Comparison of mean distribution system TTHM concentrations as a function of influent bromide concentrations

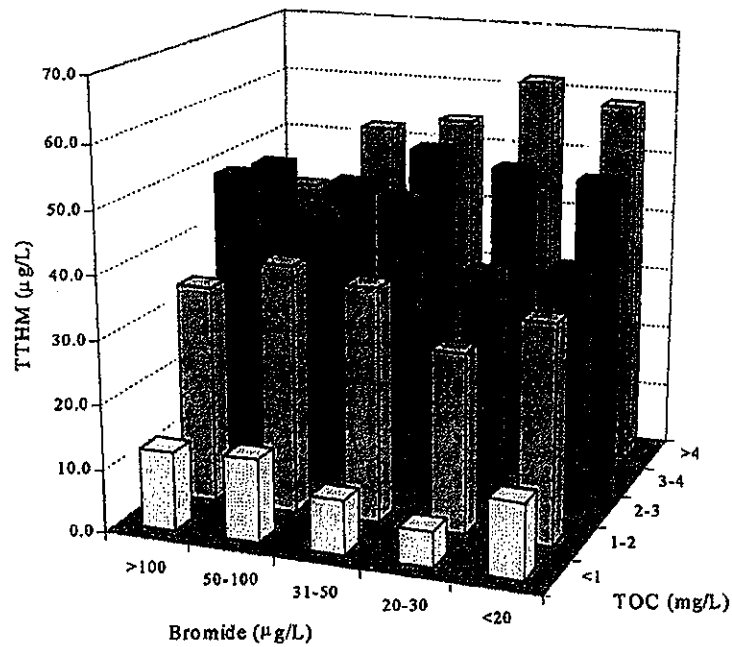


Figure 4.13 Mean distribution system TTHM concentrations plotted by influent TOC and bromide concentrations over six ICR quarters (N values were typically 150–300 for all bars)

COMPARISON OF PREVIOUS TTHM SURVEY RESULTS WITH ICR DATA

Figure 4.14 presents cumulative frequency distribution graphs for the four national TTHM surveys. Each of these surveys produced the best estimate of TTHM occurrence in U.S. drinking water at the time it was conducted. However, these four surveys took place over a 24-year period and used completely different methods of data collection. Table 4.4 summarizes the characteristics of the four national surveys, plus the limited 35-city survey of DBP occurrence. Table 4.4 shows that NORS and NOMS measured only finished water TTHM concentrations. The AwwaRF survey collected no samples and performed no independent trace chemical analyses; instead, it collected utility TTHM compliance data. The ICR TTHM data set, of course, was produced by utilities collecting samples, having them analyzed for TTHM, and then reporting the results to a central database. The ICR TTHM data that are compared with data from the other surveys described in this section consist of averages from treatment plant distribution systems and from 10 blended distribution systems.

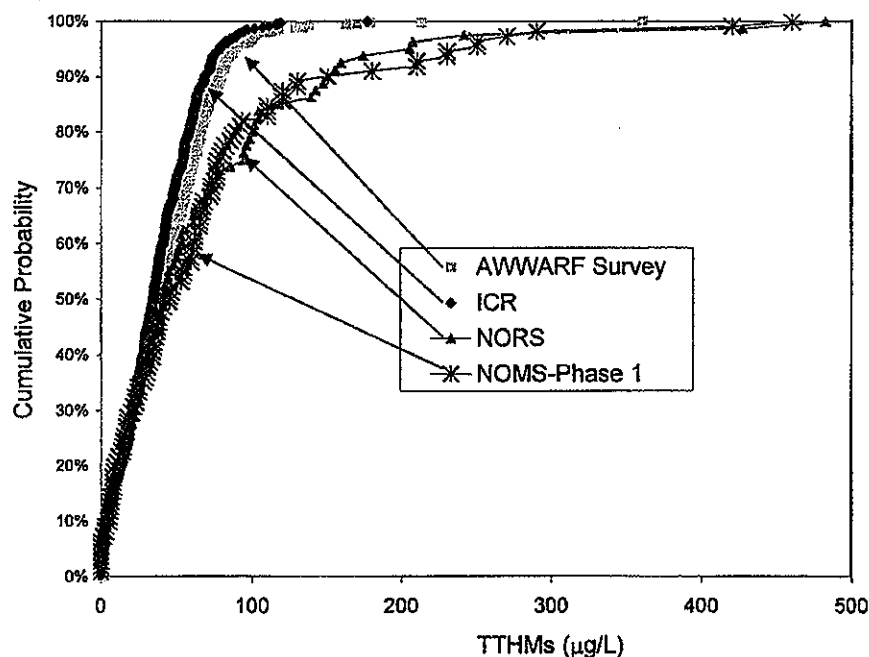


Figure 4.14 Comparison of cumulative frequency distributions from national TTHM surveys

Table 4.4
Characteristics of national TTHM surveys compared in this analysis

Characteristics	National TTHM surveys				
	NORS	NOMS Phase I	AwwaRF	35-city	ICR
Number of data points	80	111	727	35	479
Sampling period	1975	1975–76	1984–86	1988–89	1997–98
Sampling location	Plant finished water	Plant finished water	Utility distribution system (DS)	Plant finished water	469 treatment plant DSs; 10 blended DSs
Type of data	Single samples	Single samples	12 quarterly DS averages, averaged for each utility	Single samples averaged over four quarters	DS averages over six quarters
Data collection	Samples collected and shipped to USEPA lab for analysis	Samples collected and shipped to USEPA lab for analysis	Survey—samples collected by utilities and analyzed by certified labs	Samples collected and shipped to one lab for analysis	Samples collected by utility and analyzed by approved labs

Because the results of the four national TTHM surveys constitute the best estimates of national TTHM occurrence at the time they were conducted, it is possible to track changes in national TTHM concentrations over time. In Figure 4.14, the cumulative frequency distributions for NORS and NOMS data indicate much higher TTHM concentrations above a cumulative probability of about 40% compared with AwwaRF and ICR data. Below 40% cumulative probability, the data are indistinguishable from one another, because they consist of values from groundwater or surface water systems whose source water organic precursor concentrations were as low in 1997–98 as they had been in the mid-1970s.

Above 40% cumulative probability, NORS and NOMS data are similar to each other but quite different from the data collected in the AwwaRF and ICR surveys. The difference between data from NORS and NOMS on the one hand and data from the AwwaRF and ICR surveys on the other appears to be caused by utilities modifying their treatment process and disinfection practices in order to comply with the 1979 TTHM MCL. Table 4.5 lists some descriptive statistics for three national TTHM surveys and compares this information with ICR TTHM data. Data from the 35-city survey are also listed and show remarkable consistency with AwwaRF and ICR data. Table 4.5 indicates that median TTHM values for the five surveys are similar but that P75 and P90 values for the five data sets fall into two distinct groups—NORS and NOMS Phase 1 in one group and AwwaRF and ICR averages in the other.

Table 4.5
Descriptive statistics from national TTHM surveys

Survey	Number of utilities/plants	Mean (µg/L)	Median (µg/L)	P75 (µg/L)	P90 (µg/L)	Maximum (µg/L)
NORS	80	67	41	94	150	482
NOMS Phase 1	111	68	45	78	150	460
AwwaRF	727	42	39	61	79	360
ICR averages	479	36	35	54	68	177
35-city	35	45	39	52	84	185
ICR finished water	2,108*	28	23	41	60	256

* Data points

Close inspection of Figure 4.14 and Table 4.5 suggests that the 1997–98 ICR data set had slightly lower overall TTHM concentrations than the AwwaRF TTHM survey data collected during 1984–86. If there is any difference between the TTHM distributions of the two data sets, it is mainly in the area at P90 and above. If this difference is real, it suggests that reductions in national TTHM occurrence continued over the 12-year period between the two surveys, possibly in anticipation of a more stringent TTHM MCL, which was under discussion during that period. Any differences between the results of the two surveys are likely to be small and could be overwhelmed by significant differences in the manner of data collection (see Table 4.4).

As indicated in Table 4.4, the four national data sets compare finished water values with average values from distribution system sampling locations. Figure 4.15 compares the cumulative frequency distributions for ICR finished water values with the ICR averages plotted in Figure 4.14. There is a difference of 10–15 µg/L among the frequency plots for most of the probability range (excluding the very beginnings and ends of the two curves).

DISCUSSION

Because NORS and NOMS data were collected before promulgation of the TTHM rule in 1979, these data reflect only finished water values during a period when there were no limitations on the amount of chlorine added for disinfection or oxidation. The AwwaRF and ICR data sets reflect TTHM occurrence in U.S. drinking water after widespread compliance with the TTHM MCL of 0.10 mg/L (based on RAAs of TTHM data). The high end of the TTHM concentrations at the utilities represented in the AwwaRF and ICR studies is certainly lower than

the high end of the TTHM concentrations in the NORS and NOMS data sets. However, as illustrated in Figure 4.6, even the ICR survey showed very high TTHM concentrations in parts of some distribution systems.

Table 4.6 summarizes the percent reductions in finished water TTHM concentrations that occurred between NORS data and ICR data on finished water. Considering all the statistics and the relative shapes of the cumulative frequency distribution curves, a 50%–60% decrease in TTHM concentrations appears to have occurred over the 24-year period spanning NORS and the ICR.

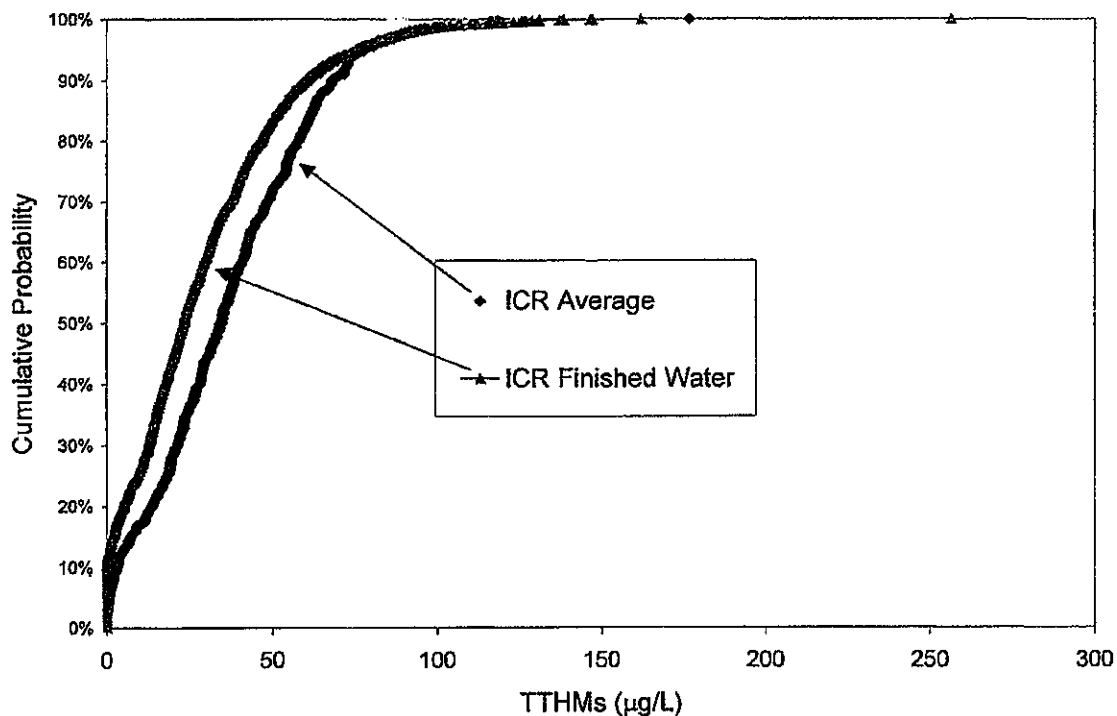


Figure 4.15 Comparison of ICR TTHM data from finished water locations and distribution system averages

Table 4.6

Percent reductions in finished water TTHM—NORS to ICR

Statistics	Percent reduction between NORS and ICR
Mean	58%
Median	44%
P75	56%
P90	60%

In 2000, the FACA committee recognized the need to reduce the high TTHM concentrations in a number of distribution systems by recommending Stage 2 DBP Rule provisions that utilities meet a TTHM MCL of 80 µg/L on average at all distribution system locations (calculated as the LRAA). If TTHM occurrence in U.S. drinking water is assessed in 2012 after full implementation of the Stage 2 DBP Rule, it would not be surprising to see a noticeable reduction in the median values, a significant reduction in the mean values, and a truly substantial reduction in the P75 and P90 values shown for ICR averages in Table 4.5. A box-and-whisker plot of this 2012 TTHM data, calculated using the LRAA concept, would very likely show no outliers or extremes above the 80 µg/L MCL.

SUMMARY AND CONCLUSIONS

Based on the data presented in this chapter, the following conclusions can be drawn:

- Four major national surveys were conducted to assess TTHM occurrence in U.S. drinking water over the period 1975 to 1998.
- In the ICR data, outlier and extreme TTHM values in distribution system samples from treatment plants using surface water or groundwater supplies indicated that a large number of TTHM values exceeded the 1979 TTHM MCL of 0.10 mg/L. These outliers and extremes significantly affected FACA negotiators, resulting in the recommendation of a Stage 2 DBP Rule provision eliminating MCL compliance through the use of RAAs.
- TTHM concentrations in ICR groundwater systems were typically far lower than TTHM values in ICR systems served by surface water. Because the majority of the outliers and extreme data points were from the distribution system samples of Florida utilities, these high groundwater TTHM values are referred to as the “Florida effect.”

- Chloroform is by far the dominant TTHM species found in drinking water samples. Four extreme values for chloroform samples collected under the ICR were near or above 300 µg/L.
- The geographic distribution of ICR TTHM data shows that the East Coast, including the southeastern states, tended to have the highest TTHM concentrations in the United States. The lowest TTHM concentrations were found in USEPA Region 10, which is composed of Oregon, Washington, Idaho, and Alaska.
- LRAAs calculated with ICR TTHM data were higher than RAAs from the same data set. Utilities will find it more difficult to comply with an 80 µg/L LRAA MCL than with an 80 µg/L RAA MCL.
- Organizing TTHM data from ICR distribution systems by type of treatment did not indicate any obvious trend except for the lower concentrations in groundwater systems. Likewise, comparing TTHM concentrations on the basis of primary or secondary disinfectant did not show tremendous differences among disinfectant categories. The four highest TTHM concentrations were from a distribution system in Mississippi that used chloramine as the secondary disinfectant.
- Correlation analyses showed that TTHM concentrations in distribution system samples were not well predicted by influent TOC concentrations alone; TTHM increased slightly with increasing TOC concentrations. High bromide concentrations in plant influent appeared to have little or no effect on distribution system TTHM concentrations. A three-dimensional plot of TOC and bromide more clearly illustrated the dependence of TTHM on TOC.
- The limited relationship between disinfectant type, source water quality, and TTHM production was primarily the result of utility efforts to aggressively treat water with high concentrations of organic and inorganic precursors in order to ensure lower TTHM concentrations and compliance with the existing MCL.
- A comparison of national TTHM occurrence surveys from 1975 to 1998 illustrated a significant decline in TTHM concentrations during this period. Four descriptive statistics for NORS and ICR finished water data indicated that TTHM concentrations have been reduced by 50%–60% over the past 24 years.

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Rule

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